Effective Thermal Conductivity of Composites Containing Spheroidal Inclusions

The effective thermal conductivity of composite materials containing perfectly-conducting, perfectly-aligned, spheroidal inclusions has been computed to $O(c^2)$, where c is the volume fraction of the inclusions, by incorporating two-particle interactions in a rigorous fashion. Relevant two-particle problems were solved by boundary collocation and method of reflections in the near and far fields, respectively. Our results for the effective thermal conductivity are rigorous extensions of Maxwell's theory in the form of an expansion in small $c\ell^{\alpha}$, where ℓ is the spheroid aspect ratio ($\alpha = 1$ for oblate and $\alpha = 2$ for prolate spheroids). The influence of microstructure was examined with two *ad hoc* model distribution functions: 1. a uniform distribution and 2. a stretched hard-sphere pair distribution. The latter yielded superior results over a greater range of c, as judged by comparisons with Willis' (1977) bounds developed from variational methods.

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Introduction

Two-phase systems find many applications in chemical engineering, such as composite materials, packed beds, fluidized beds, flow through porous media, and colloidal dispersions. The determination of the bulk properties of two-phase systems is important in optimal design of many processes and products, and thus has been of great concern to chemical engineers. For example, the design of thermally insulating materials, such as fiber-ceramic composites used on reentry space vehicles and high-performance insulation materials for buildings and storage tanks, depends upon the heat transfer characteristic of the media (Torquato, 1987). Since the late 1800's, numerous studies have been done in various two-phase problems including: the thermal conductivity, electrical conductivity, dielectric constant, magnetic permeability, and Lamé constants problems for composite materials, the mobility problem for fluidized beds, the permeability problem for porous media, and the shear viscosity problem for particle suspension systems. A unified theoretical framework for tackling these diverse problems is described in the review article by Batchelor (1974).

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The purpose of this study is to determine the relation between microstructure and the effective thermal conductivity, k_e, of an anisotropic composite material consisting of spheroidal inclusions in a homogeneous matrix. The two-phase system considered here is microscopically heterogeneous but macroscopically homogeneous: there exists a structural scale that is larger than particle dimensions and smaller than the overall dimensions of the given sample of material. By taking appropriate limits of the aspect ratio, our theory can be applied to systems ranging from fibrous composites to clay systems. Finally, results derived here apply equally well to the effective electrical conductivity, effective dielectric constant, and effective permeability problems because all are governed by the same equation.

Maxwell (1873) pioneered this approach by deriving the effective thermal conductivity of a very dilute two-phase system containing spherical inclusions. Extensions to inclusions of arbitrary shape were derived by Rocha and Acrivos (1973). Maxwell's results are correct only to $O(c)$, where c is the volume fraction of the inclusions, because particle-particle interactions are neglected. Exactly 100 years later, Jeffrey (1973) extended Maxwell's results to the $O(c^2)$ term by incorporating pair interactions. Chen and Acrivos (1976) combined the ideas in the two 1973 articles to derive the $O(c^2)$ coefficient for inclusions of slender bodies, although the exact numerical result is suspect because the interactions in nearly-touching configurations are not treated in a rigorous manner.

In general, this approach leads to a virial expansion for the thermal conductivity, strictly valid only for $c \ll 1$. In fact, as shown by Batchelor (1971) and Rocha and Acrivos (1973), the dilute theory for slender bodies requires $c\ell^2 \ll 1$, where ℓ is the particle aspect ratio, to justify in a rigorous fashion the neglect of interactions between inclusions. However, as noted by Batchelor in the same article, these restrictions are actually quite conservative. In our work, the expansions in c produce reasonable agreement with the available experimental systems over a range of volume fractions well beyond values for which the particles can be expected to remain hydrodynamically- or thermally-independent.

In another and more recent attempt to incorporate pair interactions, Hatta and Taya (1985) used the "equivalent inclusion method" to calculate the effective thermal conductivity of a randomly oriented short fiber composite. The essence of this method is to approximate inclusions by uniformly distributed thermal doublets. It is known, however, that this is only a qualitative description of pair interactions. First of all, higher-order multipole moments are required when particles are close to each other. Secondly, we note that the expression obtained by Hatta and Taya for the effective conductivity of the composite containing a equal-sized spherical inclusion is the same as Maxwell's results (which neglect particle interactions). From these, we infer that Hatta and Taya do not incorporate particle interactions properly.

In this paper, we shall examine the effect of the particle interactions on the effective thermal conductivity of the two-phase system containing spheroidal inclusions in a more rigorous manner by solving the relevant two-particle boundary value problems through the use of two numerical schemes: the method of reflections for the far field and the boundary collocation method for the near field. The accurate information of the two-particle interactions shall in turn be used to derive an expression for k_e to the $O(c^2)$ term.

We shall confine our consideration to a specific system in which the inclusion is perfectly-conducting and perfectly-aligned. This specific case is encountered commonly in such industries as the metal-enhanced plastics and graphite-enhanced plastics. The great disparity in the thermal conductivities of metals(graphites) and plastics makes the filler practically perfectly conducting compared with the matrix. For this specific composite system, the effective thermal conductivity tensor is a diagonal second-rank tensor with only two independent components: one parallel and the other perpendicular to the director of the system.

Basic Formulations

Average thermal dipole

We consider the effective thermal conductivity of a statistically homogeneous composite material for which effective properties can be defined in terms of volume-averaged quantities (Batchelor, 1974). For the present problem, the relevant quantities are the average temperature gradient $\langle \nabla T \rangle$ and the average heat flux density $\langle \boldsymbol{F} \rangle$, defined by

$$\langle \nabla T \rangle = \frac{1}{V} \int_V \nabla T \, dV = \boldsymbol{G} \tag{1}$$

and

$$\langle \boldsymbol{F} \rangle = \frac{1}{V} \int_V \boldsymbol{F} \, dV \tag{2}$$

where V is a volume large enough to contain many particles and $\boldsymbol{G}$ is the constant temperature gradient of the imposed linear ambient field $\boldsymbol{G} \cdot \boldsymbol{x}$. Because of the linearity of the governing equation, the Laplace equation, in both the matrix and particles, it follows that $\langle \boldsymbol{F} \rangle$ and $\langle \nabla T \rangle$ are linearly related,

$$\langle \boldsymbol{F} \rangle = -\boldsymbol{k}_e \cdot \langle \nabla T \rangle, \tag{3}$$

where $\boldsymbol{k}_e$ is defined as the effective thermal conductivity tensor.

The flux $\langle \boldsymbol{F} \rangle$ is usually expressed as

$$\langle \boldsymbol{F} \rangle = \frac{1}{V} \int_{V - \Sigma V_i} \boldsymbol{F} dV + \frac{1}{V} \sum_i \int_{V_i} \boldsymbol{F} \, dV$$
$$= -k_1 \langle \nabla T \rangle + n \langle \boldsymbol{S} \rangle \tag{4}$$

where n is the number density of particles and V_i denotes the volume of the ith particle. The quantity $\boldsymbol{S}$ is the thermal dipole induced by the inclusion and is defined as follows:

$$\boldsymbol{S} = \frac{k_2 - k_1}{k_2} \int_{V_p} \boldsymbol{F} \, dV = \frac{k_2 - k_1}{k_2} \int_{S_p} \boldsymbol{x} \boldsymbol{F} \cdot \boldsymbol{n} \, dA. \tag{5}$$

Here, V_p and S_p are the volume and surface of the representative particle respectively, and $\boldsymbol{n}$ is the outward unit vector normal to the particle surface. From Eq. 5, it is clear that $\langle \boldsymbol{S} \rangle$ is also linear in $\langle \nabla T \rangle$,

$$\langle \boldsymbol{S} \rangle = \boldsymbol{G} \cdot \boldsymbol{\Gamma}, \tag{6}$$

where $\boldsymbol{\Gamma}$ is a symmetric second-order tensor depending on the detail microstructure and physical properties of the system but not on the imposed ambient temperature gradient. A proof of the symmetry of $\boldsymbol{\Gamma}$ is given in Lu (1988). Thus $\boldsymbol{k}_e$ may be determined from Eqs. 3, 4 and 6 as follows:

$$\frac{\boldsymbol{k}_e}{k_1} = \delta - \frac{c}{V_p k_1} \boldsymbol{\Gamma}, \tag{7}$$

where δ is the unit tensor.

Virial expansion in c

The key quantity $\langle \boldsymbol{S} \rangle$ may be treated further as follows (O'Brien, 1979). The probability that a particle in a dilute random suspension will have m neighbors within a distance of several radii is of order c^m. If we assume that the interactions between particles fall off rapidly enough with increasing separation, then for $c \ll 1$, it seems reasonable to assume that $\langle \boldsymbol{S} \rangle$ can be calculated by a perturbation scheme; the first two terms accounting for one-body and two-body effects. In addition, we consider in this paper only systems with perfectly-aligned inclusions so that only the position distribution of the particle is involved in the averaging process. Under these circumstances, the average dipole may apparently be written as

$$\langle S \rangle = S^0 + \int_{V(R)} \{S(R|o) - S^0\} p(R|o)\, dV(R), \quad (8)$$

where $p(R|o)dV(R)$ is the probability that the center of a particle lies within the volume dV surrounding the point R, given that the center of the reference particle is at the origin. The term S^0 denotes the contribution for the dipole strength from the single-particle solution, and $\{S(r|o) - S^0\}$ is the amount by which the dipole strength of the reference particle is altered by the presence of another particle at R, i.e., the extra contribution from pair interactions.

If we proceed with higher-order interactions, then we obtain a virial expansion in c,

$$\frac{k_e}{k_1} = \delta + A_1 c + A_2 c^2 + \cdots. \quad (9)$$

Nonabsolutely, convergent integrals and renormalization

The problem of the calculation of the effective conductivity now reduces to the evaluation of the integral in the righthand side of Eq. 8. Unfortunately, the term $\{S(R|o) - S^0\}$ decays as $1/|R|^3$ as $|R| \rightarrow \infty$, and the integral is not absolutely convergent. Physically, as $|R| \rightarrow \infty$, it is incorrect to assume that the two inclusions interact through a pure matrix. Even at small (but fixed) c, we eventually encounter a separation at which screening from the other particles in the dispersion becomes significant. When this effect is taken into account, the interactions are weaker and expressible in terms of convergent integrals (Hinch, 1977).

Several approaches have been proposed to remove the difficulty of the nonabsolutely, convergent integral, for example, Batchelor's (1972) 'subtraction' procedure, Hinch's (1977) averaged-equation and renormalization approach, and O'Brien's (1979) macroscopic boundary concept. These approaches differ in appearance and ways of interpretation, but lead to the same results. Hinch's view is a particularly significant contribution, since it shows the link between the 'subtraction' method of Batchelor and the screening concepts of effective medium theory. For an overview of renormalization theory, refer to Jeffrey (1977). Here, we follow Batchelor's 'subtraction' procedure, but the same result can be obtained by more rigorous formalisms such as O'Brien's method shown in Lu (1988).

First of all, we need a 'renormalization quantity'. If we take the same consideration for $\langle \nabla T \rangle$ as for $\langle S \rangle$ in Eq. 8, we have

$$\langle \nabla T \rangle = G = (\nabla T)^0 + (\nabla T)' + O(c),$$

and thus

$$(\nabla T)^0 - G = (\nabla T)' + O(c).$$

Here, $(\nabla T)^0$ is the temperature gradient of the system neglecting particle interactions and $(\nabla T)'$ are local fluctuations. Weighting $\{(\nabla T)^0 - G\}$ with $p(R)$ and integrating over the whole space, the local fluctuations vanish (a reasonable hypothesis for systems of interest) and we find that

$$\int_{V(R)} [(\nabla T)^0 - G] p(R)\, dV(R) = O(c^2). \quad (10)$$

Here, $p(R)$ is the probability density function of finding the center of a particle at R, which, for uniformly distributed systems, is simply n, the number density of the particle. We shall show later that $[(\nabla T)^0 - G]$ is $O(1/R^3)$, which, if multiplied by an appropriate constant, can cancel the troublesome leading order term in Eq. 8 as $|R|$ tends to infinity. Our renormalzed expression for $\langle S \rangle$ is

$$\langle S \rangle = S^0 + \int_{V(R)} \{[S(R|o) - S^0] p(R|o) - M \cdot [(\nabla T)^0 - G] p(R)\}\, dV(R) + O(c^2). \quad (11)$$

The tensor M is a second rank diagonal tensor involved in the single-particle solution, which we shall address later. For spherical inclusions, Eq. 11 reduces to the expression given by Jeffrey (1973).

The $O(c)$ Coefficients

To obtain the $O(c)$ coefficients A_1 in Eq. 9, we need to solve the steady-state heat conduction problem for an isoalted ellipsoid embedded in an infinite matrix imposed with a linear ambient field $G \cdot x$. The governing equation is the Laplace equation, and the boundary conditions are continuity of the temperature fields and normal heat fluxes across the particle surface. The coordinate system is chosen so that the equation for the surface of the ellipsoid is given by

$$\frac{x^2}{a^2} + \frac{y^2}{b^2} + \frac{z^2}{c^2} = 1, \quad a \geq b \geq c,$$

where, a, b, and c are the three semiaxes of the general ellipsoid. Note that, if $b = c$ or $a = b$, the general ellipsoid reduces to the prolate spheroid and oblate spheroid, respectively.

The solution for the general ellipsoid, expressed in a form suitable for thermal dipole calculations, is available in Lu and Kim (1987):

$$T_{out}(x) = T^\infty(x) - S \cdot \nabla \int_E f_{(2)}(x') \frac{1}{4\pi k_1 |x - x'|}\, dA(x'), \quad (12a)$$

$$T_{in}(x) = T^\infty(x) - S^I \cdot x, \quad (12b)$$

and

$$S = M \cdot G, \quad (12c)$$

$$S^I = M^I \cdot G. \quad (12d)$$

Here, T_{out}, T_{in} and T^∞ denote the exterior, interior and ambient temperature fields, respectively; S is the thermal dipole generated by the presence of the particle; and M and M^I are diagonal second-rank tensors with components depending only on the geometry of the particle and the thermal conductivity of the matrix k_2 and of the particle k_1. The detail expression for the components of tensors M and M^I may be found in Lu and Kim (1987). The above solution is written in a singularity integral form. The singularity distribution function $f_{(2)}$ is a special case of the following general expression:

$$f_{(n)} = \frac{(2n-1)}{2\pi a_E b_E} q^{2n-3}, \tag{13a}$$

where

$$q(x', y') = \left[1 - \frac{x'^2}{a_E^2} - \frac{y'^2}{b_E^2}\right]^{1/2}, \tag{13b}$$

with

$$a_E = (a^2 - c^2)^{1/2}, \quad b_E = (b^2 - c^2)^{1/2}. \tag{13c}$$

The integration domain, $E(x', y')$, is the interior of the fundamental ellipse, which is the degenerate elliptical disk in a family of confocal ellipsoids, defined as follows:

$$\frac{x'^2}{a_E^2} + \frac{y'^2}{b_E^2} = 1, \quad z' = 0.$$

The $O(c)$ coefficient tensor, A_1, may be obtained by simply replacing $\mathbf{\Gamma}$ with M in Eq. 7. The result is

$$A_1 = \frac{2(k_2 - k_1)}{abc}\begin{pmatrix} C_1 & 0 & 0 \\ 0 & C_2 & 0 \\ 0 & 0 & C_3 \end{pmatrix},$$

where

$$C_i = \left[\int_0^\infty \frac{dt}{(a_i^2 + t)\Delta(t)}(k_2 - k_1) + \frac{2k_1}{abc}\right]^{-1}, \quad i = 1, 2, 3 \tag{14}$$

which is identical to the result presented in Batchelor (1974). Here a_1, a_2, and a_3 are used to denote a, b, and c, respectively. The result for prolate (oblate) spheroidal systems may be obtained by letting $b = c (a = b)$.

Pair Interactions and the $O(c^2)$ Coefficient

To obtain the $O(c^2)$ coefficients, the two-particle problem must be tackled. Again, we shall develop the solution procedure for the general ellipsoid and then reduce the results to the case of spheroids by letting $b = c$ or $a = b$.

An analytical solution of the two-ellipsoid problem is not available, thus numerical approaches are necessary. When the two particles are at large separations, the so-called method of reflections provides accurate solution with only a few iterations (Felderhof, 1977; Jeffrey and Onishi, 1984; Kim, 1985, 1986). At smaller separations, the method converges too slowly and another approach is required. Among these, the boundary collocation method has gained popularity in recent years (Gluckman et al., 1971; Liao and Krueger, 1980; Kim and Mifflin, 1985; Yoon and Kim, 1987). We shall tackle the near-field solutions by the boundary collocation method and the far-field solutions by the method of reflections. (The boundary collocation method works in all cases of interest, but the method of reflections is more efficient in the far field.)

General solution for an isolated ellipsoid

We discuss the general solution for temperature fields in and outside an isolated ellipsoid driven by an arbitrary ambient field since these are required in establishing the method of reflections and the boundary collocation method.

Our approach for an arbitrary ambient field problem is based on the expansion of an arbitrary ambient field in a Taylor series about a reference point (usually the center of the particle). Because of the linearity of the governing equation, the solution for an arbitrary ambient field may be written as a sum of solutions of subproblems consisting of the nth order ambient field, $W_{k_1k_2\ldots k_n}x_{k_1}x_{k_2}\ldots x_{k_n}$.

The analytical solution for this nth-order ambient field has been derived in Lu and Kim (1987) in the form of a distributed multipole expansion and the results are as follows:

$$T_{\text{out}}(\mathbf{x}) = T^\infty(\mathbf{x}) + \sum_{m=0}^{[(n-1)/2]} L_{(n-2m)} \int_E \frac{f_{(n-2m+1)}(\mathbf{x}')}{4\pi k_1|\mathbf{x} - \mathbf{x}'|} dA(\mathbf{x}'), \tag{15a}$$

$$T_{\text{in}}(\mathbf{x}) = T^\infty(\mathbf{x}) + \sum_{m=0}^{[(n-1)/2]} L^I_{(n-2m)} + T^c, \tag{15b}$$

where

$$L_{(n)} = \frac{(-1)^n}{n!} P_{k_1k_2\ldots k_n} \frac{\partial}{\partial x_{k_1}} \cdots \frac{\partial}{\partial x_{k_n}}, \tag{15c}$$

$$L^I_{(n)} = \frac{(-1)^n}{n!} P^I_{k_1\ldots k_n} x_{k_1} \ldots x_{k_n}, \tag{15d}$$

$$T^c = \begin{cases} D^I_{k_1\ldots k_n} W_{k_1\ldots k_n} & \text{if } n \text{ is even,} \\ 0 & \text{if } n \text{ is odd,} \end{cases} \tag{15e}$$

with

$$P_{k_1k_2\ldots k_m} = Z_{(k_1k_2\ldots k_m)(l_1l_2\ldots l_n)} W_{l_1l_2\ldots l_n}. \tag{15f}$$

The $\boldsymbol{P}$'s are the multipole moment tensors; for example, P_{k_1} are the thermal dipoles, and the $\boldsymbol{Z}$'s and $\boldsymbol{D}^I$'s are material tensors which depend only on the shape parameters (a, b, c), and the conductivities of the constituents, k_1 and k_2.

Based on Eq. 15, we find that the solution for the exterior and interior temperature fields driven by an arbitrary ambient field T^∞ may be written as

$$T_{\text{out}}(\mathbf{x}) = T^\infty(\mathbf{x}) + \sum_{n=1}^\infty P_{k_1\ldots k_n} \frac{\partial}{\partial x_{k_1}} \cdots \frac{\partial}{\partial x_{k_n}} \int_E \frac{f_{(n+1)}(\mathbf{x}')}{4\pi k_1|\mathbf{x} - \mathbf{x}'|} dA(\mathbf{x}'), \tag{16a}$$

$$T_{\text{in}}(\mathbf{x}) = T^\infty(\mathbf{x}) + \sum_{n=1}^\infty \frac{(-1)^n}{n!} x_{k_1} \ldots x_{k_n} P^I_{k_1\ldots k_n} + T^C. \tag{16b}$$

Method of Reflections

The method of reflections (MOR), first developed by Smoluchowski (1911), is essentially an iterative scheme in which the disturbance field produced by one particle is used to correct the ambient field at other particles and so forth. We confine our consideration here to two-particle systems even though this method may be applied to multiparticle systems. For two widely separated particles centered at x_1 and x_2, the zeroth-order solutions for the temperature fields are simply the sum of the disturbance solutions for each particle in isolation, i.e., without particle interactions. The zeroth-order solutions for the interior and exterior temperature fields of particle 1 (2) are denoted as $T_1^I(T_2^I)$ and $T_1(T_2)$, respectively. We note that the boundary conditions on particle 1 are violated by the extra field T_2 emanating from particle 2 and *vice versa,* and correct the situation by introducing two new exterior fields T_{21} and T_{12} and two new interior fields T_{21}^I and T_{12}^I. These new fields are obtained by taking T_2 and T_1 as ambient fields for particle 1 and 2, respectively. However, any field that corrects the boundary conditions at one particle will perturb the boundary condition at the other particle, hence we get a sequence of temperature fields comprising an iterative approximation.

At each reflection, the fields that are created to help satisfy the boundary conditions are known as *reflection fields.* The field that violates the boundary conditions (hence forcing the creation of the reflection fields) is called the *incident field.* Given an incident field, the exterior field may be represented as a multipole expansion with the moments evaluated by so-called Faxén relations (Happel and Brenner, 1983). If we consider only perfectly-conducting particle systems, the interior fields are simply constants and of no concern.

Faxén relations

When applying the method of reflections, the multipole expansion representation for the exterior field is required, i.e., a way to evaluate the associated multipole moments should be supplied. The so-called Faxén relations furnish directly such expressions for the multipole moments. The relation for ellipsoids in general thermal problems has been derived by Kim and Lu (1987) and Lu and Kim (1987). Only the results for the dipole and quadrupole moments are needed here and these may be written as:

$$S = M \cdot \int_E f_{(2)} \nabla T^{\infty}(x)|_{x=x'}\, dA, \tag{17a}$$

$$Q = \frac{1}{2} N: \int_E f_{(3)} \nabla\nabla T^{\infty}(x)|_{x=x'}\, dA, \tag{17b}$$

where N is a fourth-rank tensor whose components depend only on (a, b, c) and (k_1, k_2). The explicit expression for N may be found in Lu and Kim (1987).

$S(R|o)$ *of the first two reflections*

We consider two arbitrarily-oriented, unequal general ellipsoids in a linear ambient field $G \cdot x$. The zeroth reflection involves only the single-particle solution; from the previous discussion, we have

$$S_1^{(0)} = M_1 \cdot G, \tag{18a}$$

$$T_1 = -S_1^{(0)} \cdot \nabla \int_{E_1} \frac{f_{(2)_1} dA_1}{4\pi k_1 |x - x_1 - x_1'|}. \tag{18b}$$

In the above equations, the superscripts enclosed in parentheses denote the associated reflection numbers. The expressions for the corresponding quantities of particle 2 can be obtained by switching subscripts 1 and 2. Without the loss of generality, we shall pay attention only to particle 1.

Taking T_2 as the incident field, we may, based on Eq. 16a, write a multipole expansion solution for T_{21} as

$$T_{21} = -S_1^{(1)} \cdot \nabla \int_{E_1} \frac{f_{(2)_1}\, dA_1}{4\pi k_1 |x - x_1 - x_1'|} + \frac{1}{2} Q_1^{(1)}: \nabla\nabla \int_{E_1} \frac{f_{(3)_1}\, dA_1}{4\pi k_1 |x - x_1 - x_1'|} + \cdots, \tag{19a}$$

where

$$S_1^{(1)} = M_1 \cdot \int_{E_1} \int_{E_2} f_{(2)_1} f_{(2)_2} (M_2 \cdot G) \cdot \nabla\nabla \frac{1}{4\pi k_1 |x - x_2 - x_2'|}\bigg|_{x=x_1+x_1'} dA_2\, dA_1, \tag{19b}$$

$$Q_1^{(1)} = \frac{1}{2} N_1: \int_{E_1} \int_{E_2} f_{(3)_1} f_{(2)_2} (M_2 \cdot G) \cdot \nabla\nabla\nabla \frac{1}{4\pi k_1 |x - x_2 - x_2'|}\bigg|_{x=x_1+x_1'} dA_2\, dA_1. \tag{19c}$$

Similarly, the result for the second reflection may be constructed as

$$T_{121} = -S_1^{(2)} \cdot \nabla \int_{E_1} \frac{f_{(2)_1}\, dA_1}{4\pi k_1 |x - x_1 - x_1'|} + \frac{1}{2} Q_1^{(2)}: \nabla\nabla \int_{E_1} \frac{f_{(3)_1}\, dA_1}{4\pi k_1 |x - x_1 - x_1'|} + \cdots \tag{20a}$$

with

$$S_1^{(2)} = M_1 \cdot S_2^{(1)} \cdot \int_{E_1} \int_{E_2} f_{(2)_1} f_{(2)_2} \nabla\nabla \frac{1}{4\pi k_1 |x - x_2 - x_2'|}\bigg|_{x=x_1+x_1'} dA_2\, dA_1 - \frac{1}{2} M_1 \cdot Q_2^{(1)}: \int_{E_1} \int_{E_2} f_{(2)_1} f_{(3)_2} \nabla\nabla\nabla \cdot \frac{1}{4\pi k_1 |x - x_2 - x_2'|}\bigg|_{x=x_1+x_1'} dA_2\, dA_1 + \cdots \tag{20b}$$

The final result for the desired thermal dipoles is obtained by summing the contributions from each reflection,

$$S_i = S_i^{(0)} + S_i^{(1)} + S_i^{(2)} + \cdots, \quad i = 1, 2. \tag{21}$$

Our results may be justified as follows. We denote the distance between centers of the ellipsoids by R with $R = |z_1 - x_2|$. For two widely separated particles, $R \gg a_1$ and a_2, where a_1 and a_2 are the largest semiaxes of ellipsoid 1 and 2, respectively, we may estimate the magnitude of the greatest terms of the thermal dipoles at each reflection. From previous results, it can be seen that the greatest terms at the zeroth reflection are all $O(1)$, but those at the first reflection are $O(R^{-3})$ (These are the troublesome leading order terms in Eq. 8). The contributions become even smaller at the second reflection and are $O(R^{-6})$. This pattern persists, and we find the magnitude of the leading terms are reduced by $O(R^{-3})$ by each application of the reflection procedure. Therefore, the error becomes smaller when more reflections are included, and the result is an asymptotic solution for widely separately ellipsoids.

Given the explicit expressions for the components of material tensors $\boldsymbol{M}$ and $\boldsymbol{N}$, we can include the contributions of the multipole expansion up to the quadrupoles. The accuracy of truncation at quadrupoles may be analyzed as follows. From Eq. 20b, we see that the greatest contribution from $S_1^{(2)}$ is $O(R^{-6})$; it implies that the approximate solution for S_1 is accurate to $O(R^{-5})$ at the first reflection. Additionally, the first missing term in Eq. 20b is $O(R^{-10})$ and the greatest term in $S_1^{(3)}$ is $O(R^{-9})$. Hence, S_1 will be accurate to $O(R^{-8})$.

For oblate spheroids, the fundamental ellipse simplifies to a circle of radius a_E,

$$x'^2 + y'^2 = a_E^2.$$

The results for general ellipsoids may be applied to this case simply by using $b_E = a_E$. For prolate spheroids, the fundamental ellipse shrinks to a line segment between the two foci of the prolate spheroid and the area integral over the fundamental ellipse reduce to a line integral between the two foci:

$$\int_E f_{(n)}(x') f(x, x')\, dA(x'), \quad n \geq 2$$

$$= \frac{2n-1}{2}\frac{2n-3}{2n-2}\frac{2n-5}{2n-4}\cdots\frac{3}{4}\frac{1}{2} \cdot \int_{-1}^{1} (1-\bar{x}^2)^{n-1} f(x - a_E\bar{x}, y, z)\, d\bar{x}, \quad (22)$$

where $\bar{x} = x'/a_E$. All integrals encountered in this section may be numerically evaluated by using Gaussian quadratures (see for example Abramowitz and Stegun, 1984).

Boundary Collocation Method

In the boundary collocation method (BCM), the desired solution is written as a truncated expansion in a (complete) set of basis functions that satisfy the governing equation identically, and the unknown coefficients are determined by matching the boundary conditions at chosen points (the collocation points), thus yielding a set of linear equations for the coefficients.

Basis functions

We consider two arbitrarily-oriented, unequal-sized general ellipsoids dispersed in a continuous matrix. Based on Eq. 16a, we may propose the following basis functions:

$$T_{\text{out}} = T^\infty + \sum_{n=1}^{N} \frac{(-1)^n}{n!} P^1_{k_1\ldots k_n} \frac{\partial}{\partial x_{k_1}} \cdots \frac{\partial}{\partial x_{k_n}} \cdot \int_{E_1} \frac{f_{(n+1)_1} dA_1}{4\pi k_1 |x - x_1 - x_1'|} + \sum_{n=1}^{N} \frac{(-1)^n}{n!} P^2_{k_1\ldots k_n} \cdot \frac{\partial}{\partial x_{k_1}} \cdots \frac{\partial}{\partial x_{k_n}} \int_{E_2} \frac{f_{(n+1)_2} dA_2}{4\pi k_1 |x - x_2 - x_2'|}, \quad (23a)$$

$$T^{1I} = T^\infty + \sum_{n=1}^{N} \frac{(-1)^n}{n!} (x - x_1)_{k_1} \cdots (x - x_1)_{k_n} P^{1I}_{k_1\ldots k_n} + T^{1C}, \quad (23b)$$

$$T^{2I} = T^\infty + \sum_{n=1}^{N} \frac{(-1)^n}{n!} (x - x_2)_{k_1} \cdots (x - x_2)_{k_n} P^{2I}_{k_1\ldots k_n} + T^{2C}. \quad (23c)$$

Here, T^{1I} and T^{2I} denote the interior temperature fields for particle 1 and 2, respectively, with labels '1' and '2' used to mark the quantities associated with each particle. The exterior basis function is a multipole expansion representation of the temperature field with respect to the centers of the two particles. With this choice, $\boldsymbol{P}^1$ and $\boldsymbol{P}^2$ become the multipole moments for particle 1 and 2, respectively, and $Pa^1_{k_1}$ is the desired thermal dipole $\boldsymbol{S}(\boldsymbol{R}|\text{o})$.

We should first justify our choices. It can be seen that T_{out} satisfies the Laplace equation by noting that $\int_E f_{(n)}/|x - x'|\, dA$ (or $\int_E q^n/|x - x'| dA$) is harmonic (Lu and Kim, 1987). We also note that, when x is far away from the partlces, T_{out} approaches T^∞ with the disturbance decaying as a dipole field. The interior fields, T^{1I} and T^{2I}, also satisfy the Laplace equation if we impose the following two constraints for the $\boldsymbol{P}^{1I}$'s and $\boldsymbol{P}^{2I}$'s:

$$P^{1I}_{iik_3\ldots k_n} = 0, \quad (24a)$$

$$P^{2I}_{iik_3\ldots k_n} = 0. \quad (24b)$$

Here, the Einstein summation convention has been used to simplify the notation. These two constraints may be seen easily by taking the Laplacian of Eqs. 23b and 23c. Similar constraints as Eqs. 24a and 24b also hold for the $\boldsymbol{P}^1$'s and $\boldsymbol{P}^2$'s, but, instead of being equal to zero, the sums are some constants of no physical significance. We now have a set of basic functions for the boundary collocation method.

Before proceeding further, we discuss the number of independent variables. Without any reduction, there are 3^n components for an nth-order tensor. We note that, besides the constraints discussed above, the $\boldsymbol{P}$'s (denote as $\boldsymbol{P}^{1I}$, $\boldsymbol{P}^{2I}$, $\boldsymbol{P}^1$, $\boldsymbol{P}^2$) have permutation symmetry; for example, P_{12} is the same with P_{21}. The constraints coming from the Laplace equation and the permutation symmetry reduce the number of independent components for an nth-order tensor to only $2n + 1$. For perfectly-conducting particles, temperature gradients will not be allowed within the particles: the particles are at some uniform temperatures. Thus the $\boldsymbol{P}^{1I}$'s and $\boldsymbol{P}^{2I}$'s in the interior temperature fields are dropped. Furthermore, the number of equations obtained from each collocation point is now one instead of two, because the second boundary condition, the continuity of the heat flux density across the particle surface, is not used for perfectly-conducting particles.

The number of system variables becomes even less for equal-sized, perfectly-aligned systems. For such systems, it can be shown (Lu, 1988) that

$$P^1_{k_1\ldots k_n} = (-1)^{n+1} P^2_{k_1\ldots k_n}. \quad (25)$$

Thus, we may pick collocation points from particle 1 only and the value of T^{2C} becomes irrevelant. Considering all these constraints, the number of system variables is $(N+1)^2$ if we include, in the basis function, moments up to the 2^Nth order. Further reduction on the number of system variables is still possible when considering some specific spatial arrangements of the two particles and ambient fields. We will discuss this subject later.

The basis function for the problem of two aligned, equal-sized, perfectly-conducting general ellipsoids now take the following form:

$$T_{out} = T^\infty + \sum_{n=1}^{N} \frac{(-1)^n}{n!} P^1_{k_1\ldots k_n} \frac{\partial}{\partial x_{k_1}} \cdots \frac{\partial}{\partial x_{k_n}} \cdot \left\{ \int_{E_1} \frac{f_{(n+1)_1}}{4\pi k_1 |x - x_1 - x_1'|} dA_1 + (-1)^{(n+1)} \int_{E_2} \frac{f_{(n+1)_2}}{4\pi k_1 |x - x_2 - x_2'|} dA_2 \right., \quad (26)$$

and a constant for the interior temperature field of particle 1.

We shall now examine the basis functions for prolate and oblate spheroidal systems. For prolate spheroidal systems, we choose a set of prolate spheroidal coordinates $(\eta, \theta\ \phi)$ defined as follows:

$$\begin{cases} x = a_E \cosh\eta\cos\theta, & 0 \le \eta < \infty, \\ y = a_E \sinh\eta\sin\theta\cos\phi, & 0 \le \theta \le \pi, \\ z = a_E \sinh\eta\sin\theta\sin\phi, & 0 \le \phi < 2\pi, \end{cases}$$

where

$$a_E = \sqrt{a^2 - b^2}.$$

With this, we propose the following basis function for the prolate spheroidal system:

$$\begin{aligned} T_{out} = T^\infty &+ \sum_{n=1}^{N}\sum_{m=0}^{n} a_{nm}[Q_n^m(\zeta)P_n^m(\mu)\cos m\phi \\ &+ (-1)^{n+1} Q_n^m(\zeta')P_n^m(\mu')\cos m\phi'] \\ &+ \sum_{n=1}^{N}\sum_{m=1}^{n} b_{nm}[Q_n^m(\zeta)P_n^m(\mu)\sin m\phi \\ &+ (-1)^{n+1} Q_n^m(\zeta')P_n^m(\mu')\sin m\phi'], \end{aligned} \quad (27)$$

where $\mu = \cos\theta$, $\zeta = \cosh\eta$, the primed quantities are based on the body coordinates of particle 2, and $P_n^m(\mu)$ and $Q_n^m(\zeta)$ are the associated Legendre functions of the first and second kind, respectively, whose properties and definitions may be found in standard mathematical textbooks (for example, Hobson, 1965). In the above formulation, a_{nm} and b_{nm} are proportional to the multipole moments P^1 in Eq. 26, and the relation Eq. 25 has also been used.

Here, we need to relate the relevant dipole moments to the unknown coefficients of Eq. 27. The relation may be obtained by comparing Eqs. 26 and 27 and using Havelock's formula (Havelock, 1952):

$$P_n^m(\mu)Q_n^m(\zeta)\exp(im\phi) = \frac{1}{2}\left(\frac{\partial}{\partial y} + i\frac{\partial}{\partial z}\right)^m \int_{-a_E}^{a_E} \frac{(a_E^2 - x'^2)^{m/2} P_n^m(x'/a_E)}{|x - x'|} dx',$$

where $x' = (x', 0, 0)$. The results are as follows:

$$S_x = \frac{4\pi k_1}{3} a_E^2 a_{10},$$

$$S_y = -\frac{8\pi k_1}{3} a_E^2 a_{11},$$

$$S_z = -\frac{8\pi k_1}{3} a_E^2 b_{11}.$$

For the case of oblate spheroids, the discussion can be done in much the same way as the prolate spheroids case. First of all, we choose a set of oblate spheroidal coordinates $(\eta, \theta,]\ \phi)$ defined as follows:

$$\begin{cases} x = b_E \cosh\eta\sin\theta\cos\phi, & 0 \le \eta < \infty, \\ y = b_E \cosh\eta\sin\theta\sin\phi, & 0 \le \theta \le \pi, \\ z = \sinh\eta\cos\theta, & 0 \le \theta < 2\pi, \end{cases}$$

where

$$b_E = \sqrt{a^2 - c^2}.$$

With this, the basis function for the oblate spheroidal system becomes

$$\begin{aligned} T_{out} = T^\infty &+ \sum_{n=1}^{N}\sum_{m=0}^{n} a_{nm}[q_n^m(\zeta)P_n^m(\mu)\cos m\phi \\ &+ (-1)^{n+1} q_n^m(\zeta')P_n^m(\mu')\cos m\phi'] \\ &+ \sum_{n=1}^{N}\sum_{m=0}^{n} b_{nm}[q_n^m(\zeta)P_n^m(\mu)\sin m\phi \\ &+ (-1)^{n+1} q_n^m(\zeta')P_n^m(\mu')\sin m\phi'], \end{aligned} \quad (28)$$

where $\zeta = \sinh\eta$, $q_n^m(\zeta)$ is a special form of the associated Legendre functions of the second kind (see Lamb, 1945).

To relate the relevant dipole moments to the unknown coefficients of Eq. 28, we need the following relation between the oblate spheroidal harmonic function and the singularity integral of Eq. 26 (Lu, 1988):

$$(-1)^m \frac{(n-m)!}{(n+m)!} q_n^m(\zeta)P_n^m(\mu)\exp(im\phi) = \begin{cases} \dfrac{1}{2\pi}\displaystyle\int_E \frac{P_n^m(\cos\vartheta)\exp(im\varphi)\,dA}{p_n^m(0)b_E\sqrt{1-(x'^2+y'^2)/b_E^2}\,|x-x'|}, & p_n^m(\zeta)P_n^m(\mu)\exp(im\phi) \text{ even in } z \\ -\dfrac{1}{2\pi b_E \overline{p}_n^m(b_E)}\dfrac{\partial}{\partial z}\displaystyle\int_E \frac{P_n^m(\cos\vartheta)\exp(im\vartheta)}{|x-x'|} dA, & p_n^m(\zeta)P_n^m(\mu)\exp(im\phi) \text{ odd in } z \end{cases}$$

where

$$\bar{p}_n^m(b_E) = \lim_{\zeta \to 0} \frac{p_n^m(\zeta)}{b_E \zeta}.$$

Here, $P_n^m(\zeta)$ is a special form of the associated Legendre functions of the first kind as defined in Lamb (1945). The integration domain, $E(x', y')$, is the disk of radius b_E and may be represented by

$$\begin{cases} x' = b_E \sin\vartheta \cos\varphi, & 0 \le \varphi < 2\pi, \\ y' = b_E \sin\vartheta \sin\varphi, & 0 \le \vartheta \le \pi/2. \end{cases}$$

Using this equation and comparing Eqs. 26 and 28, we find

$$S_x = \frac{8\pi k_1}{3} b_E^2 a_{11},$$

$$S_y = -\frac{8\pi k_1}{3} b_E^2 b_{11},$$

$$S_z = -\frac{4\pi k_1}{3} b_E^2 a_{10}.$$

Application of least squares

The basis function proposed in the previous section contains in total $(N+1)^2$ unknowns, of which $N(N+1)/2 + N$ come from a_{nm}, $N(N+1)/2$ come from b_{nm}, and one from the unknown constant temperature of particle 1, T^C. If we choose $(N+1)^2$ points from the surface of particle 1 and force the basis function to satisfy the boundary condition, $T_{out}(\boldsymbol{x}_s) = T^C$, for each chosen point, then we obtain an $(N+1)^2$ by $(N+1)^2$ linear equation set. Solving this equation set, we can get the coefficients and thus the desired dipole moments. The result thus obtained is, however, quite sensitive to the location of the collocation point, and very large number of terms (this also means very high order of multipole moments) should be included to observe a significant convergence for just the first order of multipole moments, i.e., the dipoles, especially when the two particles are almost touching. We get around this problem by using a least-square approach to diminish the sensitivity of the result with respect to the locations of the collocation points. Heuristically, the information from all regions of the surface is weighted more evenly. A description of the orthogonalization method, a very accurate and efficient implementation of least squares, is given in Dahlquist and Björck (1974).

Further Treatments On Pair Interactions

For the sake of brevity, we omit the details of the numerical integration of the volume integral,

$$\int_{V(R)} [[\boldsymbol{S}(\boldsymbol{R}|o) - \boldsymbol{S}^0] p(\boldsymbol{R}|o) - \boldsymbol{M} \cdot [(\nabla T)^0 - \boldsymbol{G}] p(\boldsymbol{R})] dV(\boldsymbol{R}), \quad (29)$$

from which we obtain $\langle \boldsymbol{S} \rangle$ and then $\boldsymbol{k}_e$. Each evaluation of the integrand corresponds to the solution of the two-spheroid boundary value problem and is thus computationally-intensive. In Lu (1988), it is shown how the symmetry of the aligned-spheroid geometry may be exploited to integrate analytically over the azimuthal angle, thus reducing the volume integral to a sume of *two-dimensional* integrals. It also provides details of the numerical integration scheme (convergence, error estimates, etc.).

Results and Discussions

Our final result for the effective thermal conductivity tensor takes the form of a series expansion,

$$\frac{\boldsymbol{k}_e}{k_1} = \boldsymbol{\delta} + \left[-\frac{\boldsymbol{M}}{V_p k_1}\right] c + \left[-\frac{\boldsymbol{\Xi} + \boldsymbol{\Omega}}{V_p k_1}\right] c^2. \quad (30)$$

with Ξ and Ω defined by

$$\int_{V(R)} [\boldsymbol{S}(\boldsymbol{R}|o) - \boldsymbol{S}^0] p(\boldsymbol{R}|o)\, dV(\boldsymbol{R}) = \boldsymbol{G} \cdot \boldsymbol{\Xi} c, \quad (31a)$$

$$-\int_{V(R)} \boldsymbol{M} \cdot [(\nabla T)^0 - \boldsymbol{G}] p(\boldsymbol{R})\, dV(\boldsymbol{R}) = \boldsymbol{G} \cdot \boldsymbol{\Omega} c. \quad (31b)$$

The only thing remaining in the discussion of the evaluation of the above quantities is the effect of the probability density function $p(\boldsymbol{R}|o)$, which supplies the information on the microstructure of the two-phase medium. As discussed in Chiew and Glandt (1982), its quantitative characterization is an extremely difficult problem considering its dependence on the following factors: interparticle forces, external fields, equilibrium states, and manufacturing processes. In order to gauge the effect of microstructure, two models have been widely used by previous researchers (for systems of spherical inclusions); the *'well stirred'* model and the *hard-sphere fluid* model. In this paper, we apply the same ideas to spheroidal inclusions. We calculate the effective thermal conductivity with several different aspect ratios for prolate and oblate spheroidal systems, respectively. From these results, we determine how particle shape and system microstructure affect the particle-particle contributions to the thermal conductivity.

Well-stirred position distributions

This model assumes that particles may take nonoverlapping positions with equal probability so that the probability density function may be written as

$$\begin{cases} p(\boldsymbol{R}|o) = n, & \text{when } \boldsymbol{R} \text{ is outside of the excluded volume,} \\ p(\boldsymbol{R}|o) = 0, & \text{otherwise.} \end{cases}$$

We present the results for $\boldsymbol{k}_e/k_1$ in Table 1. We also include the results for spherical inclusions obtained by Jeffrey (1973) and the results from slender body theory of Chen and Acrivos (1976) for comparison. Our results are in agreement with Jeffrey in the limit of near spheres. In the slender-body limit, the results for the $O(c)$ coefficient are in good agreement, while the results for the $O(c^2)$ coefficient are not. The theory developed by Chen and Acrivos, as corrected by Shaqfeh, has the $O(c^2)$ coefficient as

$$A_2 = \frac{32}{315} \ell^4 \{[\ln(2\ell)]^2 [\ln(2\ell) - 1]\}^{-1},$$

but this result is based on an approximate treatment of the nearly-touching configurations. Apparently, our analysis, with

Table 1. Results for k_e/k_1 for the Well-Stirred Model, Prolate Inclusions

Aspect Ratio	$k_e^{\parallel}/k_1$	$k_e^{\perp}/k_1$
10*	$1 + 43.9c^* + 56.7c^{2*}$	
10	$1 + 49.3c + 275c^2$	$1 + 2.04c + 3.4_2c^2$
5	$1 + 17.9c + 52.9c^2$	$1 + 2.12c + 3.3_9c^2$
2	$1 + 5.76c + 10.1c^2$	$1 + 2.42c + 3.65c^2$
10/9	$1 + 3.27c + 4.97c^2$	$1 + 2.88c + 4.31c^2$
100/99	$1 + 3.02c + 4.55c^2$	$1 + 2.99c + 4.49c^2$
1.0**	$1 + 3.00c + 4.51c^{2**}$	$1 + 3.00c + 4.51c^2$

*Adopted from Chen and Acrivos (1976) as corrected by Shaqfeh (1988).
**Adopted from Jeffrey (1973).

the incorporation of more accurate (albeit numerical) results for the near-field interactions leads to larger values for the $O(c^2)$ coefficient.

The results in Table 1 show a consistent trend in A_2 except for the values of $A_2^{\perp}|_{\text{prolate}}$ for aspect ratios of 10 and 5. We see no reasons for a minimum $A_2^{\perp}|_{\text{prolate}}$ with increasing aspect ratio; instead, we believe that the flaw comes mainly from the numerical error in using BCM to solve the relevant boundary value problems when the two particles are almost touching. As expected, our results show that the prolate spheroidal system, when compared with the spherical system, gains a greater enhancement in thermal conductivity in the parallel direction while losing a relatively small amount of enhancement in the perpendicular direction. The situation for oblate spheroidal systems (Table 2) is the same except the enhanced component is in the perpendicular direction and the weakened component is in the parallel direction. This effect becomes more pronounced with increasing nonsphericity. For a given aspect ratio, the needle-like inclusion has stronger effects in enhancement than the disk-shaped inclusion. To the best of our knowledge, Table 2 contains the first results for the effective transport properties of disk-like inclusions incorporating pair interactions in a rigorous fashion.

Role of microstructure: stretched-spheres model

Apparently, the well-stirred model cannot be a good approximation for many systems because there is always some amount of potential energy existing between particles, which implies correlation between particle positions. A more reasonable choice is the hard-sphere fluid model, as used by Chiew and Glandt (1982) in their analysis of the thermal conductivity of a system of spherical inclusions. Chiew and Glandt assumed that $p(\boldsymbol{R}|\boldsymbol{o})$ is equal to the pair distribution function of the hard-sphere fluid in the equilibrium state, and found that their results fit much better with experimental data than Jeffrey's results (which was based on the well-stirred model).

Table 2. Results for k_e/k_1 for the Well-Stirred Model, Oblate Inclusions

Aspect Ratio	$k_e^{\parallel}/k_1$	$k_e^{\perp}/k_1$
10	$1 + 1.16c + 1.93c^2$	$1 + 14.4c + 58.3c^2$
5	$1 + 1.33c + 2.10c^2$	$1 + 8.02c + 19.8c^2$
2	$1 + 1.90c + 2.80c^2$	$1 + 4.23c + 7.03c^2$
10/9	$1 + 2.76c + 4.12c^2$	$1 + 3.13c + 4.74c^2$
100/99	$1 + 2.98c + 4.47c^2$	$1 + 3.01c + 4.53c^2$
1.0*	$1 + 3.00c + 4.51c^{2*}$	$1 + 3.00c + 4.51c^{2*}$

*Adopted from Jeffrey (1973).

To investigate the effect of the variation in the microstructure on the thermal conductivity of the composite, we examine an *ad hoc* position distribution function, *viz*, the stretched-spheres model of Pynn (1974) and Wulf (1977), formed by distortion of the Percus-Yevick pair distribution functions for the hard-sphere fluid (Throop and Bearman, 1965). We consider this model just to illustrate the effect of the distribution function. Other distributions, based on a more realistic incorporation of the deformation history of the composite material (unfortunately, these are as yet unavailable), are easily inserted into this formalism, since the bulk of the computations involve calculation of the portion of the integrand associated with the thermal dipole, $\boldsymbol{S}$.

The pair distribution function of the hard-sphere model fluid is a function only of the reduced radial distance r/a, i.e., may be written as $f(r/a)$. In the stretched-spheres model, the spherical coordinate is stretched (compressed) in the $x(z)$ direction to fit our prolate (oblate) spheroidal particle system. Under the new coordinate system, the reduced radial distance (now, called r/a_s) becomes $\sqrt{x^2/a^2 + (y^2+z^2)/b^2} \cdot [\sqrt{(x^2+y^2)/a^2 + z^2/c^2}]$. Replacing r/a with r/a_s, the value of the distribution function of the stretched spheres model can be evaluated readily. The ordered close packing fraction of such aligned spheroids occurs at a volume fraction of 0.7405, as discussed by Marko (1988).

In the following discussion, we denote directions along and perpendicular to the symmetry axis by superscripts $\parallel$ and $\perp$, respecitvely. The $O(c^2)$ coefficent (A_2) for the stretched-sphere model, as scaled by the corresponding result of the well-stirred distribution, is given in Tables 3 and 4, as a function of both volume fractions and spheroid aspect ratio. Plots of the effect thermal conductivity are readily obtained from these entries: Figure 1 is one such example, for the case of prolate spheroids with aspect ratio 10.

In general, the use of the stretched-sphere distribution function results in an increase in the $O(c^2)$ coefficient. Thus the predictions of the stretched-spheres model for the thermal

Table 3. A_2(Stretched-Spheres)/A_2 (Well-Stirred) for Prolate Inclusions

Aspect Ratio, c	∥ Component 10/9	2	5	10	⊥ Component 10/9	2	5	10
0.04	1.03	1.03	1.03	1.02	1.03	1.03	1.01	1.02
0.08	1.05	1.06	1.06	1.04	1.05	1.04	1.03	1.04
0.12	1.07	1.08	1.08	1.06	1.07	1.07	1.06	1.06
0.16	1.10	1.11	1.11	1.08	1.10	1.09	1.09	1.09
0.20	1.13	1.15	1.14	1.10	1.13	1.12	1.11	1.11
0.24	1.16	1.18	1.17	1.12	1.16	1.15	1.14	1.13
0.28	1.20	1.22	1.20	1.14	1.19	1.19	1.17	1.16
0.32	1.24	1.26	1.24	1.16	1.23	1.22	1.20	1.19
0.36	1.28	1.30	1.27	1.19	1.27	1.26	1.24	1.22
0.40	1.33	1.35	1.31	1.21	1.32	1.31	1.28	1.25
0.44	1.38	1.40	1.35	1.24	1.37	1.36	1.32	1.29
0.48	1.44	1.46	1.39	1.27	1.43	1.41	1.37	1.33
0.52	1.50	1.52	1.44	1.30	1.49	1.47	1.42	1.38
0.56	1.57	1.60	1.50	1.34	1.56	1.54	1.48	1.43
0.60	1.66	1.68	1.56	1.38	1.65	1.16	1.54	1.48

Table 4. A_2(Stretched-Spheres)/A_2 (Well-Stirred) for Oblate Inclusions

Aspect Ratio, c	∥ Component				⊥ Component			
	10/9	2	5	10	10/9	2	5	10
0.04	1.05	1.00	1.04	1.04	1.04	1.02	1.02	1.01
0.08	1.06	1.03	1.05	1.04	1.06	1.05	1.05	1.04
0.12	1.08	1.06	1.06	1.05	1.08	1.08	1.08	1.06
0.16	1.10	1.09	1.08	1.06	1.11	1.11	1.11	1.09
0.20	1.13	1.11	1.10	1.08	1.14	1.14	1.14	1.12
0.24	1.16	1.14	1.12	1.09	1.17	1.17	1.17	1.14
0.28	1.20	1.17	1.14	1.11	1.20	1.21	1.21	1.17
0.32	1.23	1.21	1.17	1.12	1.24	1.25	1.24	1.20
0.36	1.27	1.25	1.19	1.14	1.28	1.29	1.28	1.23
0.40	1.32	1.29	1.22	1.16	1.33	1.34	1.32	1.26
0.44	1.37	1.33	1.25	1.18	1.38	1.40	1.37	1.30
0.48	1.43	1.38	1.29	1.20	1.44	1.45	1.41	1.34
0.52	1.49	1.44	1.32	1.22	1.50	1.52	1.47	1.38
0.56	1.57	1.50	1.36	1.25	1.58	1.59	1.53	1.43
0.60	1.65	1.57	1.40	1.27	1.66	1.67	1.60	1.47

conductivity of perfectly-aligned spheroidal inclusions remain above the lower bounds derived by Willis (1977) (the upper bound diverges to infinity for prefectly-conducting systems) over a much greater range of c than the corresponding predictions using the well-stirred distribution. The lower bound developed by Torquato and Lado (1988) is also shown in Figure 1. These apply restrictions on the microstructure of aligned cylinders and does not necessarily form a lower bound for our problem. Overall, our results appear to be reasonable to fairly high-volume fractions.

Torquato (1987) gives general guidelines on the use of variational principles for the *perpendicular* component of the thermal conductivity of systems with cylindrical inclusions and notes that lower bounds often serve as a good approximation to the measurement when the difference between the properties of the two phases is extreme. These comments are borne out in Figure 1, where the predictions of the various model microstructures are shown along with the bounds and experimental data. Rather surprisingly, there is a dearth of experimental results for composites with slender inclusions of well-controlled microstructure. The experimental results of Thornburgh and Pears (1965) are for laminated composites, and thus not directly comparable to our theory, but are included in the figure to illustrate the orders of magnitudes involved.

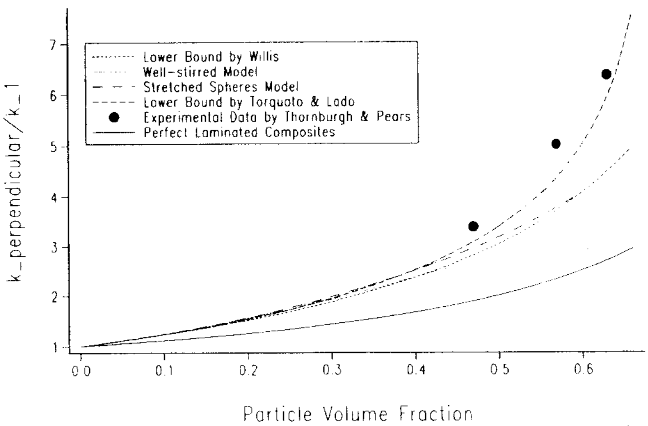

Figure 1. $k_e^{\perp}/k_1$ vs. particle volume fraction, perfectly conducting prolate spheroids with $a/b = 10$.

Comparisons with the results of Thornburgh and Pears (1965), Torquato and Lado (1988), and Willis (1977).

An examination over a range of aspect ratios reveals that $k_e^{\perp}|_{\text{oblate}}$ is less sensitive to variations in the microstructure. On the other hand, $k_e^{\parallel}|_{\text{prolate}}$ shows a relatively strong dependence on the detailed particle arrangement when the particle aspect ratio is large. This observations may be explained as follows. From Eq. 30, we see that A_2 consists of two parts, Ξ and Ω, in which only Ξ is dependent on the microstructure of the medium. In our calculation, we found that, for $k_e^{\perp}|_{\text{oblate}}$, the renormalization quantity Ω is the dominant term among the two, while for $k_e^{\parallel}|_{\text{prolate}}$, the two quantities are comparable when the aspect ratio of the particle is large. Thus, $k_e^{\parallel}|_{\text{prolate}}$ is more sensitive to variations in the microstructure. This also suggests that even greater enhancement in $k_e^{\parallel}|_{\text{prolate}}$ may be achieved by proper control of the microstructure.

Higher volume fractions: future work

As mentioned earlier, the theory developed in this paper is rigorously valid only as an expansion in the small parameter $c\ell^{\alpha}$ (with $\alpha = 1$ for oblate spheroids and 2 for prolate spheroids). However, in contrast to the behavior of the effective viscosity of concentrated suspensions, it appears that the thermal conductivity predictions of the dilute theory hold well even at very large volume fractions. We present our results to volume fractions approaching ordered closest packing [volume fraction of 0.7405 Marko (1988)] to illustrate this point.

According to percolation theory, the thermal conductivity of a composite with perfectly conducting inclusions diverges near the close-packing volume fractions. However, real dispersions of good (but not perfect) conductors at concentrations approaching these critical values still exhibit a quadratic dependence on c. The experimental result of Thornburgh and Pears is quite typical in this respect, and similar phenomena have been observed with dispersions of oblate spheroids.

A rigorous justification of these observations awaits theoretical analysis, and two directions of research seem particularly promising. The recent work of Karrila et al. (1989) suggests the possibility of tackling boundary value problems (in that paper, the Stokes equations) consisting of very many particles. The governing partial differential equation is transformed into an integral equation; the outermost eigenvalues of the relevant integral operator can be deflated to yield a contraction mapping, leading to rapid iterative solution. Indeed, compared to the Stokes equations, the deflate-and-iterate procedure for the Laplace equation is quite simple, since the spectrum requires deflation at only one end (see Kim and Karrila 1990). The recent work of Shaqfeh (1988), which follows the spirit of Hinch's (1977) renormalization theory to arrive at a 'nonlocal' analysis of the heat conduction problem, also holds promise. The synthesis of these two directions to arrive at a rigorous, yet compact, description of concentrated systems should prove to be quite worthwhile.

Conclusions

We present results for the effective thermal conductivity of an anisotropic medium composed of aligned, perfectly-conducting spheroidal inclusions. Results are given for both parallel and perpendicular components of the conductivity tensor. Rigorous values are established for the $O(c^2)$ coefficient. For prolate spheroids, our analysis spans systems ranging from near spheres to slender bodies (fibrous composites). Composites with disk-like inclusions, such as those found in ceramic systems, are also of interest as high-performance materials. Our analysis of oblate spheroids represents a significant new development for the analog of the slender body theory (the so called thin body theory) in a form suitable for handling particle-particle interactions. As far as we know, it has not been developed in the prior literature.

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Notation

a = major semiaxis of ellipsoid or spheroid
A_1, A_2 = coefficients in expansion for k_e
b = semiaxis of ellipsoid
c = minor semiaxis of ellipsoid or spheroid
C = volume fraction of dispersed phase
f = density function in singularity solution
$\boldsymbol{F}$ = heat flux
$\boldsymbol{G}$ = temperature gradient
k = thermal conductivity
ℓ; = aspect ratio
M = constant proportionality tensor in the linear relation between $\boldsymbol{S}$ and $\boldsymbol{G}$
N = constant proportionality tensor in the linear relation between $\boldsymbol{Q}$ and $\nabla\nabla T$
n = particle number density
$\boldsymbol{n}$ = surface normal
$\boldsymbol{Q}$ = thermal quadrupole
$\boldsymbol{R}$ = vector from center of particle 1 to particle 2
S = particle surface
$\boldsymbol{S}$ = thermal dipole
T = temperature
V = volume element for averages
x = position vector

Greek letters

δ = unit dyadic
η = spheroidal coordinate
θ = spheroidal coordinate
ϕ = azimuthal angle (spheroidal coordinates)

Subscripts

E = confocal ellipse
1 = particle 1
2 = particle 2
(n) = label in family of density functions

Superscripts

(n) = nth reflection
∞ = ambient field
$\perp$ = transverse tensor component
$\|$ = parallel tensor component

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