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# The Effective Thermal Conductivity of a Composite Material with Spherical Inclusions[1]

**R. H. Davis**[2]

A new method is presented for calculating the effective thermal conductivity of a composite material containing spherical inclusions. The surface of a large body is assumed kept at a uniform temperature. This body is in contact with a composite material of infinite extent having a lower temperature far from the heated body. Green's theorem is then used to calculate the rate of heat transfer from the heated body to the composite material, yielding

$$k_e/k = 1 + \frac{3(\alpha - 1)}{[\alpha + 2 - (\alpha - 1)\phi]} \{\phi + f(\alpha)\phi^2 + 0(\phi^3)\}$$

where $k_e$ is the effective thermal conductivity, $k$ is the thermal conductivity of the continuous phase, $\alpha$ is the ratio of the thermal conductivity of the spherical inclusions to $k$, and $\phi$ is the volume fraction occupied by the dispersed phase. The function $f(\alpha)$ is presented in this work. Although a similar result has been found previously by renormalization techniques, the method presented in this paper has merit in that a decaying temperature field is used. As a result, only convergent integrals are encountered, and a renormalization factor is not needed. This method is more straightforward than its predecessors and sheds additional light on the basic properties of two-phase materials.

**KEY WORDS:** composite materials; dispersions; effective properties; heat conduction; thermal conductivity.

## 1. INTRODUCTION

It is often desirable to describe a two-phase, macroscopically homogeneous material in terms of its effective bulk thermophysical properties such as

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[2] Department of Chemical Engineering, University of Colorado, Boulder, Colorado 80309, U.S.A.

shear viscosity, thermal conductivity, elastic moduli, etc. Early attempts at calculating bulk properties of a composite material or suspension containing one phase dispersed within another were beset with mathematical difficulties of a fundamental nature. Specifically, in trying to extend the classical results of Maxwell and Einstein (for the effective thermal conductivity and the effective viscosity, respectively), whose range of validity is restricted to very dilute dispersions, the early investigators encountered in their analyses nonabsolutely convergent integrals—and in some cases even divergent integrals—which appeared to render any further progress impossible. These nonconvergent integrals arose from the consideration of a representative inclusion or particle of the dispersed phase, and the summation of the pairwise interaction between this representative particle and all other particles of the dispersed phase in an infinite domain. However, techniques which avoided this difficulty and which allowed for the extension of the Maxwell and Einstein results to order $\phi^2$, where $\phi$ refers to the volume fraction of the dispersed phase, have been developed during the past decade. The most popular at present is the renormalization technique first introduced by Batchelor [1] in his analysis of the sedimentation of a statistically homogeneous, dilute suspension of monodisperse spheres. This technique has also been used to calculate the effective viscosity of suspensions [2], the effective thermal conductivity of a composite material [3], and the bulk moduli of elasticity of two-phase materials [4]. A second approach is that of Hinch [5], who constructs a hierarchy of differential equations describing the behavior of a two-phase material and shows how bulk parameters such as the sedimentation velocity can be computed in a systematic way. More recently, O'Brien [6] has computed effective properties of a dispersion by using an integral equation approach with the introduction of an artificial macroscopic boundary far from the region of interest surrounding the representative particle. O'Brien's paper [6] gives an overview of further related work.

In the present paper, a new method for predicting the effective properties of composite media is described. Although the techniques outlined in this paper may be applied to a variety of problems, we focus our attention on the effective thermal conductivity of a two-phase material containing spherical inclusions. In doing so, we consider the rate of heat transfer from a heated body placed within the two-phase material. Since the temperature field decays with the distance from the heated body, only convergent integrals are encountered in the analysis. As a result, the new method described in this paper is more straightforward than earlier techniques, and it yields additional insight into the basic properties of two-phase materials.

## 2. THEORETICAL DEVELOPMENT

Consider a body B, whose surface $A_b$ is kept at a constant temperature $T_0^*$, in contact with a composite material of infinite extent whose temperature is equal to $T_\infty^*$ far from B. The composite consists of a matrix having a thermal conductivity $k$ and spherical particles having conductivity $\alpha k$, where $\alpha$ is arbitrary. These spheres are randomly dispersed throughout the matrix and are each of radius $a$, which is assumed to be very much smaller than the linear dimensions of B. Hence, on a macroscale the composite behaves as a continuum with an effective thermal conductivity $k_e$, which is to be determined. In our theoretical development, it is assumed that $k$, $\alpha$, and $k_e$ are temperature independent. However, provided that the temperature varies significantly over a length scale that is large compared to the particle size, the final result for $k_e/k$ is independent of temperature. This restriction has already been made.

### 2.1. General Expression for the Effective Thermal Conductivity

Let $T(\mathbf{x}) \equiv [T^*(\mathbf{x}) - T_\infty^*]/(T_0^* - T_\infty^*)$ be the dimensionless temperature within the composite at any given point $\mathbf{x}$ exterior to B [with $T^*(\mathbf{x})$ being the dimensional temperature] and $T'(\mathbf{x})$ be the corresponding dimensionless temperature in the absence of spherical inclusions, i.e., the undisturbed temperature field within a solid consisting entiry of matrix material. Both $T$ and $T'$ are equal to unity on $A_b$ and vanish at infinity. In addition, $T'(\mathbf{x})$ satisfies LaPlace's equation for all points $\mathbf{x}$ outside B, with no discontinuities in its value or that of its derivatives. In contrast, $T(\mathbf{x})$ satisfies LaPlace's equation for all $\mathbf{x}$ outside B, but on the surface of each of the spherical particles its normal derivative changes by a factor of $\alpha$ when moving from the inside to the outside. Consequently, applying Green's theorem to the space occupied by the matrix material, we immediately obtain that,

$$\int_{A_b} (T\nabla T' - T'\nabla T)\cdot\mathbf{n}\,dA = (\alpha-1)\sum_{m=1}^{N}\int_{A_m} T'\nabla T\cdot\mathbf{n}\,dA \tag{1}$$

where the summation is over all $N=\infty$ spherical inclusions within the composite, $A_m$ is the surface of the spherical inclusion $m$, and $\mathbf{n}$ is the outward unit normal to the surface of the sphere. Also, in the last term of Eq. (1), the temperature gradient $\nabla T$ is evaluated on the inside of the sphere. Letting $A$ and $Q'$ be the corresponding total heat fluxes from B, i.e.,

$$Q \equiv -k(T_0^* - T_\infty^*)\int_{A_b} \nabla T\cdot\mathbf{n}\,dA \quad \text{and} \quad Q' \equiv -k(T_0^* - T_\infty^*)\int_{A_b} \nabla T'\cdot\mathbf{n}\,dA \tag{2}$$

we therefore obtain that

$$Q - Q' = k(T_0^* - T_\infty^*)(\alpha - 1) \sum_{m=1}^{N} \int_{A_m} T' \nabla T \cdot \mathbf{n} \, dA \tag{3}$$

since both $T$ and $T'$ are equal to unity on $A_b$.

Consider next a spherical inclusion with its center at $\mathbf{x}_1$. On expanding $T'$ in a Taylor series about $\mathbf{x}_1$ and rearranging the integral, we find that, for this "reference sphere,"

$$\begin{aligned} \int_{A_1} T' \nabla T \cdot \mathbf{n} \, dA &= \nabla T'(\mathbf{x}_1) \cdot \int_{V_1} \nabla T \, dV + 0(\varepsilon^5) \\ &= \frac{4\pi a^3}{3} \nabla T'(\mathbf{x}_1) \cdot \nabla T(\mathbf{x}_1) + 0(\varepsilon^5) \end{aligned} \tag{4}$$

where $V_1$ refers to the volume of the reference sphere, and $\varepsilon$ is the ratio of $a$ to the linear dimension of B and is assumed to be small. For future reference, we also note that the dipole strength of this sphere is defined in the usual manner as

$$S(\mathbf{x}_1) \equiv k(\alpha - 1) \int_{V_1} \nabla T \, dV = \frac{4\pi a^3}{3} k(\alpha - 1) \nabla T(\mathbf{x}_1) \tag{5}$$

Since for a medium of random structure the reference sphere can be centered anywhere outside $A_b'$ with a uniform probability density $3\phi/4\pi a^3$—where $\phi$ is the volume fraction of spheres and $A_b'$ encloses a region B′ consisting of B and a layer of thickness $a$ surrounding B—we can ensemble average Eq. (3) to give

$$\frac{\langle Q \rangle}{Q'} = 1 + \frac{k}{Q'} (T_0^* - T_\infty^*)(\alpha - 1)\phi \int_{\hat{V}_0} \nabla T'(\mathbf{x}_1) \cdot \nabla \langle T(\mathbf{x}_1 | \mathbf{x}_1) \rangle_1 \, d\mathbf{x}_1 \tag{6}$$

where $\hat{V}_0$ is the volume exterior to B′. In Eq. (6), use has been made of Eq. (4) with the $0(\varepsilon^5)$ term neglected. Here, $\langle Q \rangle$ is the total heat flux from B ensemble-averaged over all possible configurations of the dispersed spheres, and $\nabla \langle T(\mathbf{x}_1 | \mathbf{x}_1) \rangle_1$ is the conditional ensemble-average value of the temperature gradient at $\mathbf{x}_1$—averaged over all configurations of particles in the composite, given that there is one particle with its center fixed at $\mathbf{x}_1$; see Jeffrey [3] and Hinch [5]. It is easily seen that the right-hand side of Eq. (6) is equal to the effective thermal conductivity of the composite divided by that of the matrix material, and its definition given here is entirely consistent with the corresponding expressions given by earlier investigators.

### 2.2. Green's Theorem for the Ensemble-Average Temperature Gradient

Our next step is to apply Green's theorem using the functions $T(\mathbf{x})$ and $s^{-1}(\mathbf{x}, \mathbf{y}) \equiv |\mathbf{x} - \mathbf{y}|^{-1}$, with $\mathbf{x}$ being a fixed point anywhere in the composite, and $\mathbf{y}$ a variable point. Since $s^{-1}$ has a fundamental singularity at $\mathbf{y}=\mathbf{x}$ and is otherwise harmonic, we obtain in the usual fashion, in lieu of Eq. (1),

$$T(\mathbf{x}) = -\frac{1}{4\pi}\int_{A_b}\frac{1}{s}\nabla T\cdot\mathbf{n}\,dA - \frac{\alpha-1}{4\pi}\sum_{m=1}^{N}\int_{A_m}\frac{1}{s}\nabla T\cdot\mathbf{n}\,dA \tag{7}$$

We shall find it useful to average this equation over the ensemble of all possible configurations of spherical inclusions. This unconditional ensemble average is denoted by $\langle T(\mathbf{x})\rangle_0$. Also, we shall find the conditional ensemble average of Eq. (7)—given that there is one sphere centered at $\mathbf{x}_1$—and denote this $\langle T(\mathbf{x}\,|\,\mathbf{x}_1)\rangle_1$; similarly, the quantity $\langle T(\mathbf{x}\,|\,\mathbf{x}_1, \mathbf{x}_2)\rangle_2$ represents the average temperature at $\mathbf{x}$ over all realizations having two spheres fixed at $\mathbf{x}_1$ and $\mathbf{x}_2$, etc. Since for random dispersions there is a uniform probability density equal to $3\phi/4\pi a^3$ for spheres to be located outside B′, provided that they are centered at distances greater than $2a$ from the center of any fixed sphere, the unconditional and conditional ensemble averages of Eq. (7) are

$$\begin{aligned}\langle T(\mathbf{x})\rangle_0 = &-\frac{1}{4\pi}\int_{A_b}\frac{1}{s}\nabla\langle T(\mathbf{y})\rangle_0\cdot\mathbf{n}\,dA\\ &-\frac{3(\alpha-1)\phi}{16\pi^2a^3}\int_{\hat{V}_0}\int_{A_1}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1)\rangle_1\cdot\mathbf{n}\,dA\,d\mathbf{x}_1\end{aligned} \tag{8a}$$

$$\begin{aligned}\langle T(\mathbf{x}\,|\,\mathbf{x}_1)\rangle_1 = &-\frac{1}{4\pi}\int_{A_b}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1)\rangle_1\cdot\mathbf{n}\,dA\\ &-\frac{\alpha-1}{4\pi}\int_{A_1}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1)\rangle_1\cdot\mathbf{n}\,dA\\ &-\frac{3(\alpha-1)\phi}{16\pi^2a^3}\int_{\hat{V}_1}\int_{A_2}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1, \mathbf{x}_2)\rangle_2\cdot\mathbf{n}\,dA\,d\mathbf{x}_2\end{aligned} \tag{8b}$$

$$\begin{aligned}\langle T(\mathbf{x}\,|\,\mathbf{x}_1, \mathbf{x}_2)\rangle_2 = &-\frac{1}{4\pi}\int_{A_b}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1, \mathbf{x}_2)\rangle_2\cdot\mathbf{n}\,dA\\ &-\frac{\alpha-1}{4\pi}\int_{A_1}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1, \mathbf{x}_2)\rangle_2\cdot\mathbf{n}\,dA\\ &-\frac{\alpha-1}{4\pi}\int_{A_2}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1, \mathbf{x}_2)\rangle_2\cdot\mathbf{n}\,dA\\ &-\frac{3(\alpha-1)\phi}{16\pi^2a^3}\int_{\hat{V}_2}\int_{A_3}\frac{1}{s}\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1, \mathbf{x}_2,\ \mathbf{x}_3)\rangle_3\cdot\mathbf{n}\,dA\,d\mathbf{x}_3\end{aligned} \tag{8c}$$

where $\hat{V}_1$ is the volume exterior to B′ and to an excluded sphere of radius $2a$ centered about $\mathbf{x}_1$, and $\hat{V}_2$ is the volume exterior to B′ and to excluded spheres of radius $2a$ centered about $\mathbf{x}_1$ and $\mathbf{x}_2$. Similar equations may be written for more than two spheres fixed. We now note that the unconditional average temperature is equal to the undisturbed temperature, $\langle T(\mathbf{x})\rangle_0 = T'(\mathbf{x})$, everywhere in $\hat{V}_0$ as a result of the homogeneous nature of the composite material on a macroscopic scale. Moreover, Eq. (7) may be rederived for the function $T'(\mathbf{x})$, which is the temperature field in the absence of the dispersed phase, to yield simply

$$T'(\mathbf{x}) = -\frac{1}{4\pi}\int_{A_b}\frac{1}{s}\nabla T'\cdot\mathbf{n}\,dA \tag{9}$$

It may appear surprising that an equivalent statement cannot be made for $\langle T(\mathbf{x})\rangle_0$, in view of the fact that the two functions are equal everywhere in $\hat{V}_0$ [see Eq. (8a) for comparison]. The difference results from there being a region of inhomogeneous material of thickness $a$ surrounding the heated body B. On the inside of this thin layer, $A_b$ is in contact with matrix material only due to the exclusion effect of the spherical inclusions not overlapping with B, whereas on the outside of this layer is composite material with a uniform volume fraction $\phi$ of spheres. This nonuniformity gives rise to a jump across this layer in the normal derivative of the dimensionless temperature in proportion to $(\alpha-1)\phi$, which leads to the last term on the right-hand-side of Eq. (8a).

Since we require the spheres to be small in size compared to the heated body, fixing the position of a finite number of these spheres has a negligible effect on the normal temperature derivative at the surface of the heated body. Thus, the first terms on the right-hand sides of Eqs. (8a), (8b), and (8c) are all equal and can be evaluated by rearranging Eq. (8a). We may then let $\mathbf{x}$ be a point on the surface of the reference sphere (subscripted 1); multiplying Eq. (8) by the unit normal to this sphere and integrating over its surface yields

$$\nabla\langle T(\mathbf{x}_1)\rangle_0 = \nabla T'(\mathbf{x}_1) \tag{10a}$$

$$\begin{aligned}\frac{\alpha+2}{3}&\nabla\langle T(\mathbf{x}_1\,|\,\mathbf{x}_1)\rangle_1\\ &= \nabla T'(\mathbf{x}_1) + \frac{3(\alpha-1)\phi}{16\pi^2 a^3}\int_{\hat{V}_0}\int_{A_2}\nabla(s^{-1})\,\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_2)\rangle_1\cdot\mathbf{n}\,dA\,d\mathbf{x}_2\\ &\quad - \frac{3(\alpha-1)\phi}{16\pi^2 a^3}\int_{\hat{V}_1}\int_{A_2}\nabla(s^{-1})\,\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1,\mathbf{x}_2)\rangle_2\cdot\mathbf{n}\,dA\,d\mathbf{x}_2\end{aligned} \tag{10b}$$

$$\frac{\alpha+2}{3}\nabla\langle T(\mathbf{x}_1|\mathbf{x}_1,\mathbf{x}_2)\rangle_2$$

$$=\nabla T'(\mathbf{x}_1)-\frac{\alpha-1}{4\pi}\int_{A_2}\nabla(s^{-1})\nabla\langle T(\mathbf{y}|\mathbf{x}_1,\mathbf{x}_2)\rangle_2\cdot\mathbf{n}\,dA$$

$$+\frac{3(\alpha-1)\phi}{16\pi^2a^3}\int_{\hat{V}_0}\int_{A_3}\nabla(s^{-1})\nabla\langle T(\mathbf{y}|\mathbf{x}_3\rangle_1\cdot\mathbf{n}\,dA\,d\mathbf{x}_3$$

$$-\frac{3(\alpha-1)\phi}{16\pi^2a^3}\int_{\hat{V}_2}\int_{A_3}\nabla(s^{-1})\nabla\langle T(\mathbf{y}|\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3)\rangle_3\cdot\mathbf{n}\,dA\,d\mathbf{x}_3 \quad (10c)$$

### 2.3. Hierarchy of Integral Equations in Powers of $\phi$

In order to use Eq. (6) to calculate the heat flux from the body B—and, hence, the effective thermal conductivity of the composite material—the ensemble-averaged temperature gradient at the center of a single fixed spherical inclusion is needed. Upon rearrangement of Eq. (10b), this quantity is given by

$$\nabla\langle T(\mathbf{x}_1|\mathbf{x}_1)\rangle_1$$

$$=\frac{3}{\alpha+2}\nabla T'(\mathbf{x}_1)+\frac{9(\alpha-1)\phi}{16\pi^2(\alpha+2)a^3}\int_{\hat{V}_0-\hat{V}_1}\int_{A_2}\nabla(s^{-1})\nabla\langle T(\mathbf{y}|\mathbf{x}_2)\rangle_1\cdot\mathbf{n}\,dA\,d\mathbf{x}_2$$

$$-\frac{9(\alpha-1)\phi}{16\pi^2(\alpha+2)a^3}\int_{\hat{V}_1}\int_{A_2}(\nabla s^{-1})[\nabla\langle T(\mathbf{y}|\mathbf{x}_1,\mathbf{x}_2)\rangle_2$$

$$-\nabla\langle T(\mathbf{y}|\mathbf{x}_2)\rangle_1]\cdot\mathbf{n}\,dA\,d\mathbf{x}_2 \quad (11)$$

The first term on the right-hand side of Eq. (11) is the temperature gradient at the center of the reference sphere if it were placed alone at position $\mathbf{x}_1$ in the undisturbed temperature field, $T'(\mathbf{x})$. If the remaining two terms are neglected, which are higher order in $\phi$, then substituting Eq. (11) into Eq. (6) yields Maxwell's result as stated by Jeffrey [3]:

$$k_e/k=1+\frac{3(\alpha-1)\phi}{\alpha+2}+0(\phi^2) \quad (12)$$

In deriving Eq. (12), we have used the fact that

$$k(T_0^*-T_\infty^*)\int_{\hat{V}_0}\nabla T'(\mathbf{x}_1)\cdot\nabla T'(\mathbf{x}_1)\,d\mathbf{x}_1=Q' \quad (13)$$

which follows from Eq. (2) and application of the divergence theorem.

The second term on the right-hand side of Eq. (11) is a volume exclusion effect which discounts any configurations in the ensemble of spheres for which a second sphere would physically overlap the reference sphere. By reversing the order of integration and again applying the divergence theorem, we can show that this term is equal to $(\alpha-1)\,\phi\nabla\langle T(\mathbf{x}_1\,|\,\mathbf{x}_1)\rangle_1/(\alpha+2)$, so that Eq. (11) may be rewritten as

$$\left(1-\frac{\alpha-1}{\alpha+2}\phi\right)\nabla\langle T(\mathbf{x}_1\,|\,\mathbf{x}_1)\rangle_1=\frac{3}{\alpha+2}\nabla T'(\mathbf{x}_1)$$
$$-\frac{9(\alpha-1)\phi}{16\pi^2(\alpha+2)a^3}\int_{\hat{V}_1}\int_{A_2}\nabla(s^{-1})[\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_1,\mathbf{x}_2)\rangle_2-\nabla\langle T(\mathbf{y}\,|\,\mathbf{x}_2)\rangle_1]\cdot\mathbf{n}\,dA\,d\mathbf{x}_2 \tag{14}$$

If we now neglect the last term in this equation, we find from Eqs. (6) and (14) that

$$k_e/k=1+\frac{3(\alpha-1)\phi}{[\alpha+2-(\alpha-1)\phi]}+0(\phi^2) \tag{15}$$

which is identical to the lower bound for the effective thermal conductivity as given by the treatment of Hashin and Shtrikman [7]. It is also equivalent to the result given by Maxwell himself [8].

Equation (14) for the conditional ensemble-average temperature gradient at the center of the reference sphere is exact for all possible values of the volume fraction $\phi$. However, it is rather unwieldy because of the integral term involving the conditional ensemble-average temperature gradient with two spheres fixed, $\langle T(\mathbf{y}\,|\,\mathbf{x}_1,\mathbf{x}_2)\rangle_2$. In order to simplify this integral term, we substitute Eq. (10c) and the gradient of Eq. (8b), with $\mathbf{x}_1$ replaced by $\mathbf{x}_2$ and $\mathbf{x}$ set equal to $\mathbf{x}_1$, into Eq. (14) to yield the approximate formula

$$\left(1-\frac{\alpha-1}{\alpha+2}\phi\right)\nabla\langle T(\mathbf{x}_1\,|\,\mathbf{x}_1)\rangle_1=\frac{3}{\alpha+2}\nabla T'(\mathbf{x}_1)$$
$$+\frac{3\phi}{4\pi a^3}\int_{\hat{V}_1}\left[\nabla\langle T(\mathbf{x}_1\,|\,\mathbf{x}_1,\mathbf{x}_2)\rangle_2-\frac{3}{\alpha+2}\nabla\langle T(\mathbf{x}_1\,|\,\mathbf{x}_2)\rangle_1\right]d\mathbf{x}_2+0(\phi^2) \tag{16}$$

The error terms proportional to $\phi^2$ in Eq. (16) arise from the neglected terms proportional to $\phi$ in Eqs. (8b) and (10c). When Eq. (16) is used in Eq. (6), the error terms in the effective thermal conductivity become $0(\phi^3)$.

This technique suggests a rational method to proceed, in principle, to determine a power series in $\phi$ for the effective thermal conductivity. Namely, the $0(\phi^3)$ contribution to the power series may be obtained by retaining the terms proportional to $\phi$ when substituting Eq. (10c) and the gradient of Eq. (8b) into Eq. (14). These terms are evaluated by using Eq. (10d) (not shown) and the gradient of Eq. (8c), with the terms proportional to $\phi$ in the latter equations neglected. Higher-order terms in $\phi$ may be found by successive substitution, with the terms proportional to $\phi$ in the last equations in the substitution—those with the largest number of spheres fixed—always neglected.

## 2.4. The Expression for the Effective Thermal Conductivity Correct to $0(\phi^2)$

In order to complete our solution, the integrand of Eq. (16) must be evaluated. The term $\nabla\langle T(\mathbf{x}_1 | \mathbf{x}_1, \mathbf{x}_2)\rangle_2$ is equal to the temperature gradient at the center of a sphere located at $\mathbf{x}_1$, given that there is a second sphere centered at $\mathbf{x}_2$, with both spheres being placed in an undisturbed temperature field with gradient $\nabla T'(\mathbf{x}_1)$. This can be deduced from Eq. (10c), with the terms proportional to $\phi$ neglected as before. Similarly, $\nabla\langle T(\mathbf{x}_1 | \mathbf{x}_2)\rangle_1$ is equal [with an $0(\phi)$ correction] to the temperature field at $\mathbf{x}_1$ (where there is not a fixed sphere), given that there is a sphere centered at $\mathbf{x}_2$ in an undisturbed temperature field with gradient $\nabla T'(\mathbf{x}_1)$. These problems have been solved by Jeffrey [3], and the details are not repeated here. The key result is that

$$\nabla\langle T(\mathbf{x}_1 | \mathbf{x}_1, \mathbf{x}_2)\rangle_2 - \frac{3}{\alpha+2}\nabla\langle T(\mathbf{x}_1 | \mathbf{x}_2)\rangle_1$$

$$= -\frac{3}{\alpha+2}\sum_{p=6}^{\infty}\left(\frac{a}{r}\right)^p\left[A_p\nabla T'(\mathbf{x}_1) - B_p\frac{\mathbf{rr}}{r^2}\cdot\nabla T'(\mathbf{x}_1)\right] + 0(\phi) \tag{17}$$

where $\mathbf{r} = \mathbf{x}_2 - \mathbf{x}_1$, $r = \|\mathbf{r}\|$, and the coefficients $A_p$ and $B_p$ are known functions of $\alpha$.

Using the above result in Eq. (16) and performing the integration over the domain $\hat{V}_1$ yields

$$\left(1 - \frac{\alpha-1}{\alpha+2}\phi\right)\nabla\langle T(\mathbf{x}_1 | \mathbf{x}_1)\rangle_1$$

$$= \frac{3}{\alpha+2}\nabla T'(\mathbf{x}_1) + \frac{3\phi}{\alpha+2}\sum_{p=6}^{\infty}\frac{B_p - 3A_p}{(p-3)2^{p-3}}\nabla T'(\mathbf{x}_1) + 0(\phi^2) \tag{18}$$

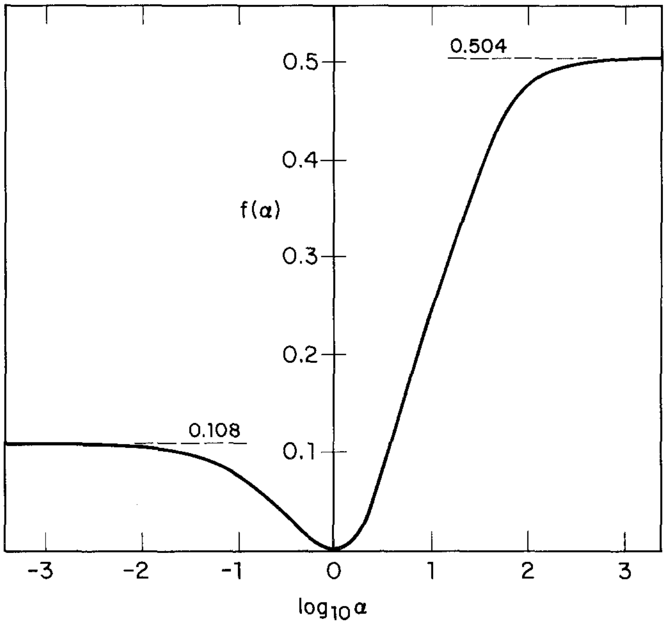


**Fig. 1.** The function $f(\alpha)$, which appears in the expression for the effective thermal conductivity given by Eq. (19).

The effective thermal conductivity may now be found from Eq. (6), which, using Eq. (13), gives our desired result:

$$k_e/k = 1 + \frac{3(\alpha-1)}{\alpha+2-(\alpha-1)\phi}\{\phi + f(\alpha)\phi^2 + 0(\phi^3)\} \tag{19}$$

where $f(\alpha) = \sum_{p=6}^{\infty} [(B_p - 3A_p)/(p-3)2^{p-3}]$, which is shown in Fig. 1.

## 3. DISCUSSION

A similar expression to Eq. (19) for the effective thermal conductivity was obtained earlier by Jeffrey [3]. This expression differs slightly from that found in the present work, but the two are in agreement to $0(\phi^2)$. In arriving at his result, Jeffrey [3] considered a composite material of infinite extent on which an undisturbed linear temperature field was imposed. As in the present work, the ensemble-averaged dipole strength of a single fixed sphere was needed, where this dipole strength is given by Eq. (5) as $\langle S(\mathbf{x}_1 | \mathbf{x}_1)\rangle_1 = 4\pi a^3(\alpha-1)\nabla\langle T(\mathbf{x}_1|\mathbf{x}_1)\rangle_1/3$. The approach taken was to equate this quantity to the dipole strength of the reference sphere in the presence of only one other sphere and then to integrate over all possible locations of the second sphere, i.e., $r \geqslant 2a$. However, Jeffrey [3] found that the pairwise correction to the dipole strength of the reference sphere due to the presence of the second sphere varied with distance as $(a/r)^3$ when the spheres are far apart. The integration of this correction over an infinite domain gives a nonabsolutely convergent integral. Jeffrey [3] therefore needed to modify his approach by introducing a renormalization quantity.

In the present approach, the undisturbed temperature gradient, $\nabla T'(\mathbf{x})$, varies inversely with the square of the distance from the heated body. Thus, the correction to the dipole strength of our reference sphere due to the presence of a second sphere varies as $(a/r)^5$, rather than $(a/r)^3$, when the two spheres are far apart. From this, we expect that the nonabsolutely convergent integral difficulties that arose when earlier workers tried to integrate the two-sphere solution will not be encountered here. Indeed, both of the integrals in Eq. (10b) are absolutely convergent and can be evaluated without rearrangement. Nonetheless, the $0(\phi^2)$ contribution to the effective thermal conductivity is not given entirely by the solution for two spheres placed in the undisturbed temperature field. Rather, the actual environment of these two spheres is the undisturbed temperature plus an $0(\phi)$ term from all of the remaining spheres. This term is given by the second term on the right side of Eq. (8a).

Our approach of averaging the integral representation of the solution of the governing equations can also be applied to problems—such as the one considered by Jeffrey [3]—where a nondecaying temperature field is applied to a composite of infinite extent. In this case, Eq. (7) for the temperature is modified by replacing the term arising from the source body B by the far field temperature $T_\infty(\mathbf{x})$ or by an integral over a macroscopic boundary far from the region of interest; see O'Brien [6]. This equation is again averaged with 0, 1, 2,... spheres fixed, and we recover Eq. (10) with $T'(\mathbf{x})$ being the applied temperature field without any spherical inclusions present. The fundamental difference is that now the integrals on the right-hand side of Eq. (10b) are no longer absolutely convergent and must be recombined as in Eq. (11). This step, however, is equivalent to the renormalization procedure used by Jeffrey [3]. The renormalization quantity would then be produced automatically by the present approach.

The method described in this paper may also be used to find the $0(\phi^2)$ contribution to other effective properties, such as elastic moduli and viscosity, for composite mdia composed of a continuous phase and dispersed spherical inclusions. The technique may also be extended, in principle, to finding higher-order contributions to these effective properties. However, even the $0(\phi^3)$ contribution requires the solution for the dipole strength of a reference sphere in the presence of two other fixed spheres. To date, a general solution of this three-sphere problem is not available. Fortunately, the higher-order contributions are not generally needed because, from comparison with exact solutions for ordered arrays of spherical inclusions [9], Eq. (19) for the effective thermal conductivity is accurate to within a few percent for all possible values of $\alpha$ and $\phi$, except in the dual limit $\alpha \to \infty$ and $\phi \to \phi_{\max}$, where $\phi_{\max} \approx 0.62$ is the maximum random packing volume fraction with the spheres touching one another. In this limit of densely packed, perfectly conducting spheres, the effective thermal conductivity approaches infinity logarithmically and may be estimated from the formulas by Batchelor and O'Brien [10] or Sangani and Acrivos [9].

## REFERENCES


1. G. K. Batchelor, *J. Fluid Mech.* **52**:245 (1972).
2. G. K. Batchelor and J. T. Green, *J. Fluid Mech.* **56**:461 (1972).
3. D. J. Jeffrey, *Proc. R. Soc.* **335**:355 (1973).
4. H. S. Chen and A. Acrivos, *Int. J. Solids Struct.* **14**:349 (1978).
5. E. J. Hinch, *J. Fluid Mech.* **83**:695 (1977).
6. R. W. O'Brien, *J. Fluid Mech.* **91**:17 (1979).
7. Z. Hashin and S. Shtrikman, *J. Appl. Phys.* **33**:10, 3125 (1962).
8. J. C. Maxwell, *Electricity and Magnetism* (Clarendon Press, London, 1873).
9. A. S. Sangani and A. Acrivos, *Proc. R. Soc. Lond.* **A386**:263 (1983).
10. G. K. Batchelor and R. W. O'Brien, *Proc. R. Soc. Lond.* **A355**:313 (1977).