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## THERMAL CONDUCTION IN COMPOSITES

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### ABSTRACT

We consider the effective thermal conductivity of two-component isotropic composites and review bounds obtained through analytic continuation of the effective conductivity as a function of the component conductivities. The connection between this conductivity function and Stieltjes functions is emphasized. Many of the well-known bounds on the effective thermal conductivity correspond to bounds on Stieltjes functions and these bounds, in turn, are closely related to Padé approximants.

### INTRODUCTION

The emphasis of this paper is different from that of the conference presentation. Here we review analytic properties of the effective thermal conductivity,  $\bar{\lambda}_0$ , of composites, rather than proving that the well-known effective-medium approximation [1,2] is exact for a specific class of model composites. These model composites, which have a self-similar or fractal-like character, are described elsewhere [3].

The effective conductivity of a statistically isotropic and homogeneous two-component composite is defined as the constant of

proportionality,  $\bar{\lambda}_0$ , in the equation

$$\{J_T(\underline{x})\}_\Omega = -\bar{\lambda}_0 \{VT(\underline{x})\}_\Omega \quad (1)$$

relating the average of the heat current  $J_T(\underline{x})$  to the average of the temperature gradient  $\nabla T(\underline{x})$  in the composite. The brackets are used to denote averages, in this case an average over a large cubic volume,  $\Omega$ , of the composite. The fields  $J_T(\underline{x})$  and  $T(\underline{x})$  are often calculated by solving the microscopic equations of thermal conductivity:

$$J_T(\underline{x}) = -\lambda(\underline{x})\nabla T(\underline{x}), \quad \nabla \cdot J_T(\underline{x}) = 0, \quad (2)$$

where the local conductivity  $\lambda(\underline{x})$  takes two values:  $\lambda_1$  in component 1 and  $\lambda_2$  in component 2. Our work is based on these equations. To justify their use we make four simplifying assumptions. First, we suppose the components are in good thermal contact so that there is no temperature discontinuity across interfaces. Second, the temperature must be sufficiently high to ensure that the mean free path of phonons or free electrons is much smaller than the characteristic size of inhomogeneities. Third, the temperature must be low enough to neglect heat transport due to radiation and convection. Last, we suppose the cross-coupling between different fields is negligible. (This is reasonable when the thermopowers and thermoelastic coefficients are small.)

Our overall aim is to reproduce known bounds on the effective conductivity that have practical applications and then provide an elegant generalization of them. We start by reviewing the well-known connection between thermal conductivity in composites and other transport processes. We then discuss a perturbation solution for  $\bar{\lambda}_0$ , which is useful when the structure is arbitrary and  $\delta\lambda \equiv \lambda_1 - \lambda_2$  is small. Finally, following Bergman [4,5] and others [6-11] analytic properties of  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  are studied and used to obtain bounds on the effective conductivity. We draw attention to the connection between these bounds and the bounds on Stieltjes functions derived by Baker [12,13].

#### RELATED EFFECTIVE CONSTANTS

Electrical conductivity in a composite is described by the equations

$$J_E(\underline{x}) = -\sigma(\underline{x})\nabla V(\underline{x}), \quad \nabla \cdot J_E(\underline{x}) = 0, \quad (3)$$

where  $J_E(\underline{x})$  is the electrical current,  $V(\underline{x})$  the electric potential and  $\sigma(\underline{x})$  is the local electrical conductivity taking values  $\sigma_1$  in

component 1 and  $\sigma_2$  in component 2. The similarity between (2) and (3) implies that the effective electrical conductivity  $\bar{\sigma}_0(\sigma_1, \sigma_2)$  is precisely the same function of  $\sigma_1$  and  $\sigma_2$  as  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  is of  $\lambda_1$  and  $\lambda_2$ , for a given composite with fixed structure. Similarly the effective dielectric constant  $\bar{\epsilon}_0(\epsilon_1, \epsilon_2)$  is the same function of  $\epsilon_1$  and  $\epsilon_2$ . (See the remarks of J. D. Patterson in these proceedings.) Hence if the ratios  $\sigma_1/\lambda_1$  and  $\sigma_2/\lambda_2$  are equal, then  $\bar{\sigma}_0/\lambda_0$  also shares the same value. This implies that if the Wiedemann-Franz law [14] applies to both component materials, then it works for the composite as well. The similarity does not extend to the effective elastic constants of composites, which relate average stress to average strain fields. Nevertheless, simple correlations between the thermal conductivity and elastic properties of composites have been established [3].

#### A PERTURBATION SOLUTION

One approach to estimating  $\bar{\lambda}_0$  using a perturbation scheme is due to Brown [15], who deduced formal expressions for the coefficients,  $y_m$ , in the series expansion,

$$\bar{\lambda}_0 = \{\lambda\}_0 + \delta_\lambda \sum_{m=1}^{\infty} y_m (\delta_\lambda / \{\lambda\}_0)^m, \quad (4)$$

where

$$\delta_\lambda \equiv \lambda_1 - \lambda_2, \quad \{\lambda\}_0 \equiv f_1 \lambda_1 + f_2 \lambda_2, \quad (5)$$

in which  $f_1$  and  $f_2 = 1 - f_1$  are the volume fractions occupied by components 1 and 2. Brown related  $y_m$  to a multiple integral over an  $(m + 1)$  - point correlation function which gives the probability that all  $m + 1$  points lie in component 1 (or 2). In the special case  $m = 1$ , this integral is independent of the structure of the composite and Brown proves  $y_1 = -f_1 f_2 / d$ , where  $d$  is the dimensionality of the composite. The next important contribution to  $\bar{\lambda}_0$  comes from the term involving  $y_2$ . This coefficient can be expressed [15,16] in the form

$$y_2 = f_1 f_2 [f_2 + (d-1)\zeta_1 - d f_1] / d^2 = -f_1 f_2 [f_1 + (d-1)\zeta_2 - d f_2] / d^2, \quad (6)$$

where, for a three-dimensional composite, we define

$$\zeta_1 \equiv 1 - \zeta_2 = \frac{9}{2f_1 f_2} \int_0^\infty dr \int_0^\infty ds \int_{-1}^{+1} du \frac{f_{111}(r, s, u)}{rs} P_2(u), \quad (7)$$

in which  $P_2(u)$  is a Legendre polynomial and  $f_{111}(r,s,u)$  is the probability that all three vertices of a given triangle lie in component 1; the triangle having sides  $r$ ,  $s$  and included angle  $\cos^{-1}u$ . This fundamental geometric parameter  $\zeta_1$  has been calculated for a variety of composites, including cell materials [17] (described below), regular arrays of spheres [18], and systems of both penetrable spheres [19] and hard spheres [20] randomly inserted in a matrix. The simplest results are for the cell materials, constructed by dividing space into cells and then flipping the same weighted coin in each cell: heads, which occurs with probability  $f_1$ , means the cell is to be filled with component 1; tails means it is to be filled with component 2. Miller [17], who devised these materials, proves  $\zeta_1 = f_1$  for spherical cells,  $\zeta_1 = f_2$  for platelike cells, and  $\zeta_1 = (f_2 + 3f_1)/4$  for needlelike cells. The results have been generalised to spheroidal cells of arbitrary eccentricity [18]. For cells to be considered truly platelike, the aspect ratio of the equatorial axis to the longitudinal axis must exceed 100. For needle-like cells, this aspect ratio must be smaller than 1/10.

In the following section, the coefficients  $y_m$  in the expansion (4) are incorporated in bounds on  $\bar{\lambda}_0$ . These bounds often give reliable estimates for the effective conductivity. Note that the series expansion is expressible in various equivalent forms through a change of variables. For instance,  $1/\bar{\lambda}_0$  can be expressed as a series in terms of

$$\delta_{1/\lambda} \equiv 1/\lambda_1 - 1/\lambda_2, \quad \{1/\lambda\}_0 \equiv f_1/\lambda_1 + f_2/\lambda_2, \quad (8)$$

having the same basic form as (4), but with different coefficients.

#### ANALYTIC PROPERTIES AND BOUNDS

The function  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  has some very beautiful properties first recognized by Bergman [4] and rigorously proved by Golden and Papanicolaou [10]. Specifically the function can be expressed in the form

$$\bar{\lambda}_0(\lambda_1, \lambda_2) = a_0\lambda_1 + b_0\lambda_2 + \lambda_1g_0(\lambda_1/\lambda_2), \quad (9)$$

where  $a_0$  and  $b_0$  are real-valued satisfying

$$a_0 \geq 0, \quad b_0 \geq 0, \quad a_0 + b_0 \leq 1, \quad (10)$$

and  $g_0(z)$  has the integral representation

$$g_0(z) = \int_0^\infty \frac{d\phi_0(u)}{1+zu}, \quad (11)$$

in which  $\phi_0(u)$  is a bounded non-decreasing function normalized to ensure

$$\bar{\lambda}_0(1, 1) = 1. \quad (12)$$

The representation (11) is, in fact, the defining equation for a Stieltjes function. The properties of Stieltjes functions have been extensively studied in the mathematics literature; Baker [13] gives an excellent review. Evidently from (11),  $g_0(z)$  is an analytic function of  $z$  with singularities restricted to the negative real  $z$  axis. These singularities include simple poles with positive residues, essential singularities and branch cuts. For example, Bergman [21] and McPhedran and McKenzie [22] consider regular arrays of identical spheres (and cylinders) of conductivity  $\lambda_1$  in a matrix of conductivity  $\lambda_2$ . Their numerical work indicates that for these composites  $g_0(z)$  has an essential singularity at  $z = -1$  which forms the accumulation point of a set of poles located along the negative real axis. In any composite with some degree of randomness, the poles are likely to get smeared out and form a branch cut [4]. Indeed, Dykhne [23] proves  $\sqrt{\lambda_1 \lambda_2}$  is the exact effective conductivity of any two-dimensional, possibly random, composite that is symmetric in the two-components, like a checkerboard. This implies  $g_0(z) = 1/\sqrt{z}$ , with a branch cut along the entire negative real axis.

Qualitative features of the function  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  can sometimes be determined from measurements of the effective dielectric constant taken over those frequencies where the wavelength greatly exceeds the characteristic size of inhomogeneities. (At higher frequencies the radiation is scattered.) The dielectric constants  $\epsilon_1$ ,  $\epsilon_2$  and  $\bar{\epsilon}_0$  are then generally complex with imaginary parts that govern the absorption of incident radiation. For example, in the cermet Ag - SiO<sub>2</sub> the silica has a real and relatively constant dielectric dielectric constant,  $\epsilon_2$ , whereas the dielectric constant,  $\epsilon_1$ , of silver varies with frequency along a trajectory in the upper half of the complex plane. This trajectory passes very close to the negative real  $\epsilon_1$  axis, i.e., near where the singularities of  $g_0(z)$  are expected to have the greatest influence. The measurements of  $\bar{\epsilon}_0$  with  $f_1 = 0.39$ , taken by Abeles and Gittleman [24] exhibit a pronounced absorption peak at a wavelength of about 0.45 microns. This resonant absorption, not present in either pure silver or pure silica indicates the presence of a pole (or short branch cut singularity) in  $g_0(z)$ . Physically it is due to the same sort of resonance occurring in capacitor-inductor networks: the silica acts as a capacitor and the silver serves as an inductor.

Although the analytic properties of  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  have been known since the pioneering work of Bergman [4], the connection with Stieltjes functions has been recognized only recently. In fact, many of the bounds on  $\lambda_0$  deduced from first principles by Bergman [4,5], Felderhof [8] and one of us (GWM) [6,7] could have

been directly obtained from the bounds on Stieltjes functions derived by Baker [12]. These bounds incorporate the coefficients in the series expansion (4) up to any given order and are closely related to Padé approximants [13]. To see how they arise, we follow the approach of Baker [12], also adopted by others [11,42]. For simplicity, let us suppose  $\lambda_1 > \lambda_2$ . First note that the inequality

$$\frac{1}{z} \int_0^\infty \frac{d\phi_0(u)}{1+u} \leq g_0(z) \leq \int_0^\infty \frac{d\phi_0(u)}{1+u}, \quad \text{for all } z > 1, \quad (13)$$

which follows from (11), can be combined with (9), (10) and (12) to establish the elementary bound,

$$\lambda_2 \leq \bar{\lambda}_0 \leq \lambda_1. \quad (14)$$

Now the invariance properties of Stieltjes functions imply that if  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  has the series expansion (4) with  $y_1 = -f_1 f_2 / d$ , then, recalling the definitions (5) and (8),

$$\bar{\lambda}_1(\lambda_1, \lambda_2) \equiv (d-1)^{-1} \lambda_1 \lambda_2 (\bar{\lambda}_0 \{1/\lambda\}_0 - 1) / (\{\lambda\}_0 - \bar{\lambda}_0) \quad (15)$$

is a function of the same character as  $\bar{\lambda}_0(\lambda_1, \lambda_2)$ , i.e. it can be expressed in the form

$$\bar{\lambda}_1(\lambda_1, \lambda_2) = a_1 \lambda_1 + b_1 \lambda_2 + \lambda_1 g_1(\lambda_1/\lambda_2), \quad (16)$$

where  $g_1(z)$  is a Stieltjes function, and  $a_1$  and  $b_1$  are non-negative with sum  $a_1 + b_1 \leq 1$ . The normalization constant  $(d-1)^{-1}$  in (15) serves to ensure  $\bar{\lambda}_1(1, 1) = 1$ . Since all the terms in (16) contributing to  $\bar{\lambda}_1$  are positive when  $\lambda_1, \lambda_2 > 0$ , it follows that  $\bar{\lambda}_1 > 0$  and through (15) this implies

$$\{1/\lambda\}_0^{-1} \leq \bar{\lambda}_0 \leq \{\lambda\}_0. \quad (17)$$

These bounds, established by Wiener in 1912 using an entirely different method [25], were the best available for 50 years. Hashin and Shtrikman [26] made the next major advance. They discovered new variational principles (incorporating trial "polarization" fields) which yielded an improved set of bounds. By analogy with (14) we have

$$\lambda_2 \leq \bar{\lambda}_1 \leq \lambda_1 \quad (18)$$

which in conjunction with (15) yields bounds on  $\bar{\lambda}_0$  that are in fact equivalent to the bounds of Hashin and Shtrikman. They prove there exist composites [26] which attain these bounds. The lower bound in (18) is attained when the composite is an assemblage of

spheres of component 1 each coated with a shell of component 2 such that the components are in the same proportion,  $f_1:f_2$ , in every coated sphere. In order to fill all space the coated spheres must have a variety of sizes, ranging to the infinitesimal. The upper bound corresponds to a similarly constructed material where the roles of the components are interchanged. Hence (18) are the best possible bounds that incorporate  $f_1$  (or  $f_2$ ) and no other information about the isotropic composite. For a comparison with experiment, see Corson [27] and DeVera and Strieder [28]. Recently, Tartar and Murat [29] and Lurie and Cherkaev [30] have generalized these bounds to anisotropic composites, using the method of compensated compactness. Their bounds are attained when the composite is an aggregate of coated ellipsoids that are aligned and fill all space.

Note that (4) and (6) when substituted in (15), imply

$$\bar{\lambda}_1(\lambda_1, \lambda_2) = \{\lambda\}_1 + O(\delta_\lambda^2) = \{1/\lambda\}_1^{-1} + O(\delta_\lambda^2), \quad (19)$$

where

$$\{\lambda\}_1 \equiv \zeta_1 \lambda_1 + \zeta_2 \lambda_2, \quad \{1/\lambda\}_1 \equiv \zeta_1/\lambda_1 + \zeta_2/\lambda_2. \quad (20)$$

Hence the geometric parameters  $\zeta_1$  and  $\zeta_2 = 1 - \zeta_1$ , defined by (7), now play a similar role to the volume fractions  $f_1$  and  $f_2 = 1 - f_1$ . By analogy with (17) we deduce the bounds

$$\{1/\lambda\}_1^{-1} \leq \bar{\lambda}_1 \leq \{\lambda\}_1. \quad (21)$$

which are identical with bounds due to Beran [31] when expressed in terms of  $\bar{\lambda}_0$ . [Beran's bounds have been considerably simplified by Torquato and Stell [32] and Milton [16]: the connection with (21) becomes evident only after this simplification]. Few experimental tests of these bounds have been made owing to the difficulty in measuring the three point correlation function  $f_{111}(r,s,u)$  needed to evaluate  $\zeta_1$  via (7) [27]. If, however, the elastic properties of the composite are known, then this provides an alternative method for estimating  $\zeta_1$ . The bounds on the effective bulk and shear moduli of composites deduced by Beran and Molyneux [33], McCoy [34] and Milton and Phan-Thien [35], in fact, incorporate  $\zeta_1$  [16].

An entire sequence of bounds can be constructed by generalizing the preceding arguments [11,12]. We introduce a hierarchy of conductivity functions  $\bar{\lambda}_j(\lambda_1, \lambda_2)$  defined for  $j = 1, 2, \dots$ , in terms of  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  through the recursion relation

$$\bar{\lambda}_{j+1}(\lambda_1, \lambda_2) \equiv n_j \lambda_1 \lambda_2 (\bar{\lambda}_j \{1/\lambda\}_j - 1) / (\{\lambda\}_j - \bar{\lambda}_j), \quad (22)$$

where the normalization constant  $n_j > 0$  is chosen to ensure  $\bar{\lambda}_{j+1}(1, 1) = 1$  and the averages

$$\{\lambda\}_j \equiv w_{1,j}\lambda_1 + w_{2,j}\lambda_2, \quad \{1/\lambda\}_j \equiv w_{1,j}/\lambda_1 + w_{2,j}/\lambda_2, \quad (23)$$

incorporate non-negative weights  $w_{1,j}, w_{2,j}$  defined by

$$w_{1,j} \equiv 1 - w_{2,j} \equiv \left. \frac{\partial \bar{\lambda}_j(\lambda_1, 1)}{\partial \lambda_1} \right|_{\lambda_1=1}. \quad (24)$$

The normalization constants  $n_j$  and weights  $w_{1,j}$  can be expressed, using (22), in terms of the coefficients  $y_m$  in the series expansion (4). They respectively depend on  $2j + 2$  and  $2j + 1$  point correlation functions. From (4), (15) and (20) we have

$$n_0 = (d - 1)^{-1}, \quad w_{1,0} = f_1, \quad w_{1,1} = \zeta_1. \quad (25)$$

In the special case where  $\bar{\lambda}_0(\lambda_1, \lambda_2)$  is a rational function of  $\lambda_1$  and  $\lambda_2$ , the hierarchy terminates when  $\bar{\lambda}_j = \{\lambda\}_j$  or  $\{1/\lambda\}_j^{-1}$ : beyond this the conductivity functions are clearly not defined.

As  $j$  increases, the inequalities

$$\lambda_2 \leq \bar{\lambda}_j \leq \lambda_1, \quad \{1/\lambda\}_j^{-1} \leq \bar{\lambda}_j \leq \{\lambda\}_j \quad (26)$$

imply successively tighter bounds on  $\bar{\lambda}_0$  which include progressively more information about the composite. The nested sequence of bounds generated by the first pair of inequalities are known as even-order bounds, since they depend on correlation functions up to an even order,  $2j$ . The second pair of inequalities generate a nested sequence of odd-order bounds.

Common [36] and Baker [12] obtain the corresponding sets of bounds on Stieltjes functions. The problem was first suggested by Common who deduced a hierarchy of inequalities, later sharpened (and extended to complex  $\lambda_1/\lambda_2$ ) by Baker. Beran [31] and Kröner [37] describe how odd-order and even-order bounds up to arbitrarily high order can be deduced from variational principles. Their bounds coincide with the bounds on  $\bar{\lambda}_0$  implied by (26), which in turn are equivalent to the bounds in Refs. 7, 9 and 11 [38]. This generalizes the work of Bergman [4], who established that the Wiener [25] and Hashin-Shtrikman [26] bounds follow from the analytic properties of



$\bar{\lambda}_0(\lambda_1, \lambda_2)$ ; a conclusion reached in the above analysis.

Beran [31] first raised the question of whether the bounds converge as  $j \rightarrow \infty$ . The answer is yes, provided  $\lambda_1/\lambda_2$  is not zero, infinite, or real and negative. Indeed, as Baker establishes in his monograph [13], the moment problem is determinate for a Stieltjes series with a non-zero radius of convergence. McPhedran and Milton [18] calculate series expansions for the conductivity of regular arrays of nearly touching spheres of conductivity  $\lambda_1$  in a matrix having conductivity  $\lambda_2$ . They find the bounds on  $\bar{\lambda}_0$  converge rapidly as  $j$  increases for conductivity ratios  $\lambda_1/\lambda_2$  up to 10,000. In fact, the width of these bounds is negligible when  $j \geq 7$ . In this sense, the normalization constants  $n_j$  and the weights  $w_{1,j}$  characterize  $\bar{\lambda}_0(\lambda_1, \lambda_2)$ . To calculate them is in general a tedious and difficult task, requiring knowledge of high-order correlation functions which are not usually available. Two exceptions are worth noting. First, for symmetric materials we have  $\bar{\lambda}_0(\lambda_1, \lambda_2) = \bar{\lambda}_0(\lambda_2, \lambda_1)$ . This implies  $w_{1,j} = w_{2,j} = \frac{1}{2}$  for all  $j$ . Second, Keller's identity [39],

$$\bar{\lambda}_0(1/\lambda_1, 1/\lambda_2) = 1/\bar{\lambda}_0(\lambda_1, \lambda_2), \quad (27)$$

which holds for any two-dimensional (isotropic) composite implies  $n_j = 1$  for all  $j$ . When  $d > 2$ , (27) takes the form of an inequality, derived by Schulgasser [40], and the normalization constants  $n_j$  for  $j \geq 1$  all depend on the structure of the composite. The effective medium approximation [1,2] for an aggregate of spheres corresponds to a choice of parameters  $n_j = (d-1)^{-1}$  and  $w_{1,j} = f_1$  for all  $j$  [41].

The above analysis extends to anisotropic composites. For these composites, the effective thermal conductivity is represented by a  $d$ -dimensional symmetric matrix  $\bar{\lambda}_0$  with non-negative eigenvalues  $\lambda_{0,k}$ ,  $k = 1, 2, \dots, d$ . For simplicity, let us suppose the structure of the composite remains unchanged under spatial reflection, which ensures that the eigenvectors (or principal axes) of  $\bar{\lambda}_0$  do not rotate as the ratio  $\lambda_1/\lambda_2$  varies. Then any eigenvalue, as a function of  $\lambda_1$  and  $\lambda_2$ , is expressible in the familiar form [4,6,10],

$$\bar{\lambda}_{0,k}(\lambda_1, \lambda_2) = a_{0,k}\lambda_1 + b_{0,k}\lambda_2 + \lambda_1 g_{0,k}(\lambda_1/\lambda_2) \quad (28)$$

where the positive constants  $a_{0,k}$  and  $b_{0,k}$  have sum at most 1 and  $g_{0,k}(z)$  is a Stieltjes function. Furthermore  $\bar{\lambda}_{0,k}(\lambda_1, \lambda_2)$  has a series expansion in  $\delta\lambda$  and  $\{\lambda\}_0$ , of the form (4) with new coefficients  $y_{m,k}$  ( $m = 1, 2, \dots, \infty$ ). The leading coefficients are correlated through the identity [6]

$$\sum_{k=1}^d y_{1,k} = -f_1 f_2. \quad (29)$$

In conjunction with the representation (28) this identity implies bounds on  $\lambda_0$  that are precisely equivalent to the ones obtained by Tartar and Murat [29] and Lurie and Cherkaev [30]. To see this it is necessary to introduce functions  $\bar{\lambda}_{1,k}(\lambda_1, \lambda_2)$  each related to  $\lambda_{0,k}(\lambda_1, \lambda_2)$  through an equation analogous to (22).

One would hope that similar considerations apply to the effective thermal conductivity of multicomponent composites and polycrystalline materials. Despite significant progress [4,7,11], this remains an outstanding problem deserving more attention.

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