Effective Conductivity of Dispersions of Spheres with a Superconducting Interface

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We consider the problem of exactly determining the effective thermal conductivity, σ_{eff}, of a composite material consisting of periodic or random arrays of spheres which possess superconducting interfaces. This requires the solution of the local temperature field in which there is a jump in the normal component of the heat flux across the sphere–matrix interface. The strength of this interface is characterized by a dimensionless parameter $C \geqslant 0$ such that $C = 0$ corresponds to the usual perfect interface case. An approximate formula for the effective conductivity, σ_{eff}, compares favourably with our results for a wide range of sphere volume fractions, phase conductivity ratios, and values of the interfacial strength C.

1. Introduction

In an earlier paper (Cheng & Torquato 1997), henceforth referred to as paper I, we discussed the effect of interfacial resistance on the effective conductivity of periodic arrays of spheres using an analytical method. In the present work, we will study the effect of an imperfect interface that exhibits superconductance. A superconducting interface is tantamount to one in which there is a jump in the normal component of the heat flux, although the temperature is continuous across the interface. This model was proposed and analysed using variational bounding methods by Torquato & Rintoul (1995). These authors used classical minimum energy principles to find very sharp, rigorous bounds on the effective properties of a class of dispersions with superconducting interfaces (characterized by a dimensionless parameter $C \leqslant 0$) that incorporates crucial microstructural information about the interface. It was shown that there exists a critical value, C_{c}, above which non-conducting inclusions (relative to the matrix) can actually increase the effective conductivity above that of the matrix conductivity.

   In this paper, we will determine analytically the effective conductivity of periodic arrays of spheres with superconducting interfaces using Rayleigh's method. We will also compute the effective conductivity of random arrays with superconducting interfaces using an integral equation approach. Both of these methods have been proven to be able to provide accurate results for conductivity calculations in the perfect interface case (McPhedran & McKenzie 1978; McKenzie et al. 1978; Sangani & Acrivos 1983; Hinson & Felderhof 1992; Greengard & Moura 1994). To make our results as accurate as possible, we have incorporated contributions from all multipole terms to a very high order to insure that we have fully resolved the problem to spectral accuracy. In the periodic case, we have used an efficient and accurate linear

system solver from LINPACK, while in the random case the iterative GMRES was employed to solve the discretized linear system. The lattice summations involved in our calculations were obtained accurately using the method described in Berman & Greengard (1994). The non-zero lattice sums for order $n \leqslant 20$ were tabulated in the Appendix of paper I.

The remainder of the paper is organized as follows. In §2, we describe in detail the boundary-value problem for the temperature field and the solution techniques for periodic and random arrays of spheres with superconducting interfaces. We derive also an exact formula for the effective conductivity of such periodic arrays through order $O(f^9)$, where f is the volume fraction of the spheres. In §3, we present all of our numerical results for periodic and random arrays and some discussion. In §4, we show that an approximation expression for the effective conductivity that incorporates a three-point microstructural parameter (see Torquato & Rintoul 1997) predicts the effective conductivity remarkably well.

2. Formulation and solution technique

In this section, we will summarize the formulation of the effective conductivity problem for arrays of spherical inclusions with a superconducting interface. The solution techniques for both periodic and random arrays are discussed.

(a) Formulation of the problem

We consider a composite material consisting of periodic or random arrays of equal-sized spherical inclusions of radius a and conductivity σ_2 embedded in a uniform matrix of conductivity σ_1. Following Torquato & Rintoul (1997), it is assumed that there exists a superconducting layer of zero thickness between the matrix and included phases. The strength of this superconductance is characterized by a dimensionless parameter C, adopting the notation from Torquato & Rintoul (1997), which can be defined through a limiting process by first considering a three-phase composite in which each inclusion possesses a concentric coating of thickness δ and conductivity σ_c and then letting $\delta \to 0$ and $\sigma_c \to \infty$ such that

$$C = (\tilde{C}/\sigma_1 a), \quad \text{with} \quad \tilde{C} = \lim_{\substack{\delta \to 0 \\ \sigma_c \to \infty}} \delta \sigma_c. \tag{2.1}$$

Torquato & Rintoul (1997) showed that such a superconducting interface leads to a jump in the heat flux across the interface, as opposed to a temperature jump as in the resistance case. In fact, for a superconducting interface the temperature is now continuous across the interface. The jump in the normal component of the flux is related to the temperature at the interface through the relation

$$T_- = T_+ = (a^2/2\tilde{C})[J_n^- - J_n^+], \tag{2.2}$$

where T_+ and T_- are the temperatures as one approaches the inclusion surface from the matrix and particle side, respectively, and

$$J_n^+ = -\sigma_1 \frac{\partial T_+}{\partial n}, \quad J_n^- = -\sigma_2 \frac{\partial T_-}{\partial n}$$

are the normal flux from the matrix and particle sides, respectively.

Therefore, if we apply a uniform field of strength E_0 along the negative z-axis to the composite material, the harmonic temperature field generated in the material

shall satisfy the following boundary-value problem inside the cubic unit periodic cell Ω with sides of length 1:

$$\nabla \cdot (\sigma \nabla T) = 0, \quad \text{in } \Omega, \tag{2.3}$$

$$\sigma(\boldsymbol{x}) = \begin{cases} \sigma_2, & \text{if } \boldsymbol{x} \in V, \\ \sigma_1, & \text{if } \boldsymbol{x} \in \Omega \backslash V, \end{cases}$$

$$T_- = T_+ = \frac{a}{2\sigma_1 C} \left[\sigma_1 \frac{\partial T_+}{\partial n} - \sigma_2 \frac{\partial T_-}{\partial n} \right] \quad \text{on } \partial V, \tag{2.4}$$

with periodic boundary conditions given by

$$\begin{aligned} T(x+1, y, z) &= T(x, y, z), \\ T(x, y+1, z) &= T(x, y, z), \\ T(x, y, z+1) &= T(x, y, z) - E_0. \end{aligned} \tag{2.5}$$

Here $\boldsymbol{x} = (x, y, z)$ is a position vector, V is the volume occupied by the spherical inclusion, ∂V is the sphere surface, and $\partial/\partial n$ is the outward normal derivative on ∂V. Thus our problem differs from both the perfect interface case and the interfacial resistance case at the boundary condition (2.4). In the perfect interface case, both the temperature and the normal component of the flux are continuous across the interface. In the interfacial resistance case, the normal component of the flux is continuous but the temperature jumps across the interface.

Equations (2.3)–(2.5) will be solved using a Rayleigh-like method for both the periodic case and the random case, although for the random case the equation is established through an integral equation. Once the temperature field is obtained, the effective conductivity of the composite material can be calculated via the homogenized relation

$$\langle \boldsymbol{J} \rangle = \boldsymbol{\sigma}_{\text{eff}} \langle \boldsymbol{E} \rangle, \tag{2.6}$$

where $\boldsymbol{\sigma}_{\text{eff}}$ is generally the second-order effective conductivity tensor. Under the assumption of isotropy, it is characterized by a single scalar parameter σ_{eff} through the relation $\boldsymbol{\sigma}_{\text{eff}} = \sigma_{\text{eff}} \boldsymbol{I}$, where $\boldsymbol{I}$ is the 3×3 unit matrix. The average intensity, $\boldsymbol{E}$, and the average flux, $\langle \boldsymbol{J} \rangle$, are given by

$$\langle \boldsymbol{E} \rangle = \frac{1}{V} \int_V \nabla T \, \mathrm{d}V + \frac{1}{V} \int_{\partial V} a \left(\frac{\partial T_+}{\partial n} - \alpha \frac{\partial T_-}{\partial n} \right) \boldsymbol{n} \, \mathrm{d}S, \tag{2.7}$$

$$\langle \boldsymbol{J} \rangle = \frac{1}{V} \int_V \boldsymbol{J} \, \mathrm{d}V, \tag{2.8}$$

where $\alpha = \sigma_2/\sigma_1$. Note that the average potential gradient, $\boldsymbol{E}$, in (2.7) now contains an additional integral involving the normal flux jump across the interface ∂V, in comparison with those for perfect interface and with interfacial resistance case. It can be shown that

$$\langle \boldsymbol{J} \rangle = \sigma_1 \left[\langle \boldsymbol{E} \rangle - (1 - \alpha) \int_{\partial V} \boldsymbol{x} \frac{\partial T_-}{\partial n} \, \mathrm{d}S - 2C \int_{\partial V} T_- \boldsymbol{n} \, \mathrm{d}S \right],$$

upon using the same techniques as we used in Cheng & Torquato (1997), i.e. using the multipole expansion of temperature field, we were able to obtain,

$$\sigma_{\text{eff}} = \sigma_1 \left(1 - \frac{4\pi B_{1,0}^*}{E_0} \right),$$

where $B_{1,0}^*$ is the total induced dipole of the periodic cell. This relation is true for all configurations.

(b) Solution technique: periodic case

We consider periodic arrays of identical spheres centred on the sites of simple, body-centred and face-centred cubic lattices. The choice of unit cell and the related lattice sums remain the same as in paper I (Cheng & Torquato 1997).

The potential in the matrix will be represented by

$$T_1(r, \theta, \phi) = \sum_{n=0}^{\infty} \sum_{m=-n}^{n} \left(A_{nm} r^n + \frac{B_{nm}}{r^{n+1}} \right) Y_n^m(\theta, \phi), \tag{2.9}$$

and the potential inside the sphere by

$$T_2(r, \theta, \phi) = \sum_{n=0}^{\infty} \sum_{m=-n}^{n} C_{nm} r^n Y_n^m(\theta, \phi), \tag{2.10}$$

where $Y_n^m(\theta, \phi)$ are spherical harmonics. With these representations, and the well-known Rayleigh's field identity, equations (2.3)–(2.5) can be solved in terms of the coefficients A_{nm}, B_{nm} and C_{nm}. The resulting infinite-dimensional linear system for B_{nm} is given as follows:

$$\frac{B_{nm}}{a^{2n+1}} \frac{m+1+m\alpha+2C}{m-m\alpha-2C} = E_0 \delta_{n1} \delta_{m0} + \sum_{i=0}^{\infty} \sum_{j=-i}^{i} (-1)^i J_m^j \mathcal{L}_{n+i}^{j-m} \Upsilon_{n+m}^{i-j} \Upsilon_{n-m}^{i+j} B_{ij}, \tag{2.11}$$

for $n = 0, 1, \ldots, \infty$, $m = -n, \ldots, n$. The reader is referred to paper I for details to derive this system. In (2.11), δ_{ij} is the standard Kronecker delta symbol, $\mathcal{L}_n^m$ are three-dimensional lattice sums defined by

$$\mathcal{L}_n^m = \sum_{p \in \Lambda_3} Y_n^m(\theta_p, \phi_p)/r_p^{n+1}, \quad n \geqslant 1, \quad m = -n, \ldots, 0, \ldots, n, \tag{2.12}$$

where Λ_3 denotes the set $\{(k_1, k_2, k_3) \mid k_i \in \mathbf{Z}, (k_1, k_2, k_3) \neq (0, 0, 0)\}$, and

$$J_m^{m'} = \begin{cases} (-1)^{\min(|m'|, |m|)}, & \text{if } m \cdot m' > 0, \\ 1, & \text{otherwise}, \end{cases} \tag{2.13}$$

$$\Upsilon_k^l = \sqrt{\binom{k+l}{k}}, \tag{2.14}$$

for any integer m, m', k, l.

The infinite linear system, (2.11), will be truncated and solved numerically using the direct method for all three cubic lattices. Due to the full symmetry of cubic arrays, simplifications of (2.11) can be sought to greatly reduce the number of unknowns. In fact, in (2.11), with our choice of the unit cells (see paper I), only B_{nm} with n odd and m being exactly multiples of four are non-zero. Discarding those vanishing B_{nm} can significantly ease the computational effort.

Remark 2.1. An exact solution for the effective conductivity can be obtained from (2.11) through order $O(f^9)$ just as in paper I for the resistance case; the resulting

formula remains unchanged and is given by

$$\frac{\sigma_{\text{eff}}}{\sigma_1} = 1 - \frac{3f}{D},$$ (2.15)

in which D is

$$D = -\beta_1^{-1} + f + c_1\beta_3 f^{10/3} + c_2\beta_5 f^{14/3} + c_3\beta_3^2 f^{17/3} + c_4\beta_7 f^6 + c_5\beta_3\beta_5 f^7$$
$$+ c_6\beta_9 f^{22/3} + O(f^{25/3}),$$

with constants $c_1, \ldots, c_6$ for three cubic lattice arrays tabulated in table 1 of paper I. However, β_i is now defined by the relation

$$\beta_i = \frac{i\alpha - i + 2C}{i\alpha + i + 1 + 2C}.$$ (2.16)

(c) Solution technique: random case

Let us now consider a random distribution of M identical spherical inclusions of radius a in the computational cell. This system of M spherical inclusions is then replicated periodically in all directions to fill the entire space. It is straightforward to extend the method described above for the periodic case to such configurations. However, to solve the resulting linear equation system, we would have to use an iterative method instead of the direct-solution approach because the order of the linear system is very large. Unfortunately, the iterative method does not converge quickly when applied to linear systems obtained by the previous method. Thus, we turn to a different formulation of the problem based on the Fredholm integral equation of the second kind. It is well known that the numerical solution of such an integral equation is very stable. For the perfect interface case, this formulation was studied in detail by Cheng (1995). For our superconducting-interface case, a change in the integral equation is necessary, but most of the details remain the same. We will give a brief review of the method here, paying particular attention to the modifications due to the superconducting interface (see Cheng (1995) for further details).

To begin with, the temperature in the computational cell is now represented as a single layer potential:

$$T(\boldsymbol{x}) = -E_0 z + \sum_{\text{images}} \sum_{i=1}^{M} \int_{\partial V_i} G(\boldsymbol{x}, \boldsymbol{y}) \rho_i(\boldsymbol{y}) \, \mathrm{d}s_{\boldsymbol{y}},$$ (2.17)

where $G(\boldsymbol{x}, \boldsymbol{y}) = -(1/4\pi|\boldsymbol{x} - \boldsymbol{y}|)$ is the free-space Green's function for the Laplacian, z is the z-component of $\boldsymbol{x}$, $\rho_i(\boldsymbol{y})$ represents the induced surface charge density which remains to be determined and $\mathrm{d}s_{\boldsymbol{y}}$ is an infinitesimal surface element at point $\boldsymbol{y}$ on the surface of the inclusions. The summation over *images* takes into account all images of our computational cell in the periodic tiling of the entire space.

It is clear that the solutions represented by (2.17) satisfy equation (2.3) and the periodic boundary conditions, together with the continuity condition across the interface. Hence, the only condition that remains to be fulfilled is the jump condition

$$T_-(\text{or } T_+) = \frac{a}{2\sigma_1 C} \left[\sigma_1 \frac{\partial T_+}{\partial n} - \sigma_2 \frac{\partial T_-}{\partial n} \right],$$ (2.18)

at each inclusion interface. Substitution of (2.17) into this condition yields the

following Fredholm integral equation of the second kind for $\rho_j(x)$:

$$\rho_j(x) - 2\lambda \sum_{\text{images}} \sum_{i=1}^{M} \int_{\partial V_i} \frac{\partial G}{\partial n_x}(x,y)\rho_i(y)\,ds_y - \frac{4C}{a(1+\alpha)} \sum_{\text{images}} \sum_{i=1}^{M} \int_{\partial V_i} G(x,y)\rho_i(y)\,ds_y$$

$$= 2\lambda E_0 \frac{\partial z}{\partial n_x} + \frac{4C}{a(1+\alpha)} E_0 z, \quad \text{for } x \in V_j, \qquad (2.19)$$

for $j = 1, 2, \ldots, M$, where $\lambda = (\sigma_2 - \sigma_1/\sigma_2 + \sigma_1)$ is a contrast parameter. Relative to the corresponding perfect-interface expression, we see that (2.19) involves two extra terms related to the parameter C. To solve this equation numerically, we can expand the surface charge density $\rho_j(x)$ in terms of spherical harmonics:

$$\rho_j(x) = \sum_{n=0}^{\infty} \sum_{m=-n}^{n} A_{nm}^j Y_n^m(\theta, \phi), \qquad (2.20)$$

for $j = 1, 2, \ldots, M$, where (a, θ, ϕ) are the spherical coordinates of x with respect to the centre of V_j. The coefficients $\{A_{nm}^j\}$ for $n = 0, 1, 2, \ldots, \infty$ and $m = -n, \ldots, n$ are unknowns to be determined. The development of the linear system for $\{A_{nm}^j\}$ is an application of classical potential theory (see Cheng 1995 for details).

3. Results and discussion

In this section we present numerical results using the methods described in the previous section. For periodic configurations, results are reported for simple, body-centred and face-centred cubic lattice cases, using the same numerical tools as in paper I. For the random case, the linear system is solved using a complex version of the popular iterative method GMRES of Saad & Schultz (1986).

(a) Periodic case

We first present the results for simple cubic lattice arrays. Table 1 lists the dimensionless effective thermal conductivity $\sigma_{\text{eff}}/\sigma_1$ for a conductivity ratio $\sigma_2/\sigma_1 = 0.1$ and a case of virtually perfectly insulating inclusions in which $\sigma_2/\sigma_1 = 0.00001$. In each case, we give results for two different values of the dimensionless conductance parameter, C, and a wide range of volume fractions. Figures 1 and 2 depict the results for $\sigma_2/\sigma_1 = 0.1$ and $\sigma_2/\sigma_1 = 0.00001$, respectively, as a function of the sphere volume fraction f. The perfect interface results ($C = 0$) are also included in the figures.

As in the case of interfacial resistance, the results show that the interfacial superconductance also has a dramatic effect on the effective property. However, whereas interfacial resistance reduces the effective conductivity, interfacial superconductance increases the effective property. In the superconducting case, the critical value of the dimensionless parameter C for a given conductivity ratio α, is $C_c = \frac{1}{2}(1 - \alpha)$ (Torquato & Rintoul 1995). At this critical value, the inclusions are again effectively 'hidden', i.e. the effective conductivity σ_{eff} is exactly equal to the matrix conductivity σ_1. When $C > C_c$, it is seen that the superconducting interface can make relatively insulating inclusions behave effectively as conducting inclusions.

Tables 2 and 3 give corresponding results for body-centred and face-centred cubic arrays, respectively. Qualitatively, these results are similar to those for the simple cubic array illustrated in figures 1 and 2 and hence are not depicted graphically.
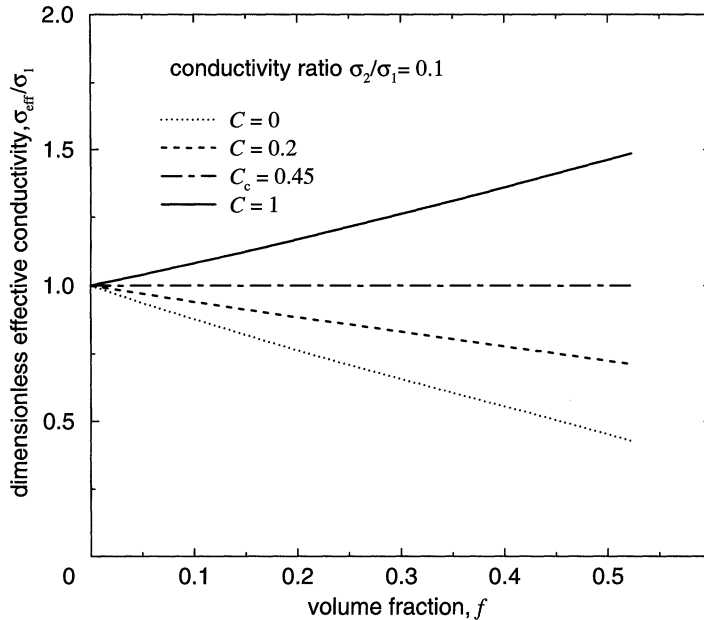
Figure 1. Dimensionless effective conductivity $\sigma_{\text{eff}}/\sigma_1$ versus volume fraction f for a simple cubic lattice with sphere to matrix conductivity ratio $\alpha = \sigma_2/\sigma_1 = 0.1$. The dotted line is the previously-known result for the perfect-interface case ($C = 0$). The dashed line is for a moderate interfacial conductance ($C = 0.2$), the dash-dotted line is for the critical interfacial conductance value ($C_c = 0.45$), and the solid line is for a large interfacial conductance ($C = 1$).

Figure 2. Dimensionless effective conductivity $\sigma_{\text{eff}}/\sigma_1$ versus volume fraction f for a simple cubic lattice with sphere to matrix conductivity ratio $\alpha = \sigma_2/\sigma_1 = 0.00001$. The dotted line is the previously-known result for the perfect-interface case ($C = 0$). The dashed line is for a moderate interfacial conductance ($C = 0.2$), the dash-dotted line is for the critical interfacial conductance value ($C_c = 0.499\,995$), and the solid line is for a strong interfacial conductance ($C = 1000$).
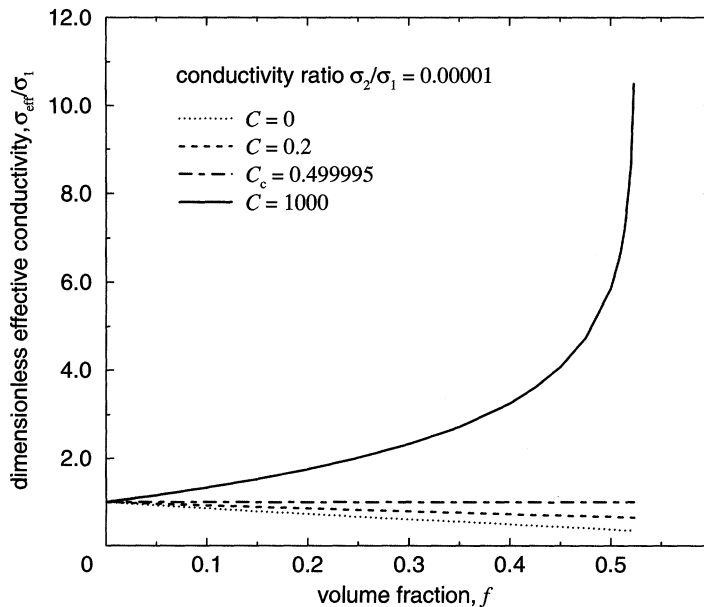
Table 1. *Dimensionless effective thermal conductivity $\sigma_{\mathrm{eff}}/\sigma_1$ of simple cubic lattice of spherical inclusions with imperfect interfaces versus the volume fraction f*

(Here $f_{\max} = \frac{1}{6}\pi$. The critical value $C_c = 0.45$ for $\alpha = 0.1$ and $C_c = 0.499\,995$ for $\alpha = 0.000\,01$.)

	conductivity ratio $\alpha = 0.1$		conductivity ratio $\alpha = 0.000\,01$	
f	$C = 0.2$	$C = 1$	$C = 0.2$	$C = 1000$
0.000	1.000 00	1.000 00	1.000 00	1.000 00
0.050	0.970 30	1.040 79	0.962 96	1.157 66
0.100	0.941 17	1.082 71	0.926 82	1.333 00
0.150	0.912 60	1.125 78	0.891 53	1.529 95
0.200	0.884 55	1.170 07	0.857 02	1.754 46
0.250	0.856 97	1.215 59	0.823 21	2.015 94
0.300	0.829 81	1.262 38	0.790 01	2.330 03
0.350	0.803 01	1.310 46	0.757 29	2.724 62
0.400	0.776 48	1.359 83	0.724 90	3.256 38
0.425	0.763 28	1.384 99	0.708 77	3.611 34
0.450	0.750 12	1.410 47	0.692 64	4.071 40
0.475	0.736 95	1.436 25	0.676 45	4.723 93
0.500	0.723 74	1.462 32	0.660 11	5.850 86
0.510	0.718 44	1.472 83	0.653 50	6.695 04
0.515	0.715 78	1.478 11	0.650 17	7.379 68
0.520	0.713 12	1.483 39	0.646 82	8.607 06
0.523	0.711 52	1.486 56	0.644 79	10.5033

Finally, to illustrate the jump in the normal component of the flux across the superconducting interface graphically, we present in figure 3 surface plots of the quantity $\sigma(x)\partial T/\partial r$ in the plane $y = 0$ and $z = 0.25$ for the simple cubic lattice case for the conductivity ratio $\alpha = 0.1$ and volume fraction $f = 0.5$. The two rows of figure 3 correspond to $C = 0$ (top) and $C = 1$ (bottom), respectively. The results for body-centred and face-centred cubic arrays are similar and hence are not plotted.

(b) *Random case*

The effective conductivity for random suspensions of spheres with a perfect interface has been studied by Kim & Torquato (1991) using the first-passage-time (Brownian motion) method and by Bonnecaze & Brady (1991) using a simulation method featuring exact two-body interactions. The exact numerical calculation for large random systems for arbitrary values of volume fractions and phase properties is very difficult due to prohibitive convergence and storage problems. On the other hand, periodic arrays of spheres with perfect interfaces have been treated very successfully by McPhedran & McKenzie (1978), McKenzie *et al.* (1978) and Sangani & Acrivos (1982) by exact methods.

Here we will present results obtained through exact numerical calculations described in §2c. We have been able to do so with the help of the three-dimensional fast multipole method (Greengard 1985). We have treated problems with a cell size

Table 2. *Dimensionless effective thermal conductivity σ_{eff}/σ_1 of body-centred cubic lattice of spherical inclusions with imperfect interfaces versus the volume fraction f*

(Here $f_{max} = (\sqrt{3}/8)\pi$. The critical value $C_c = 0.45$ for $\alpha = 0.1$ and $C_c = 0.499995$ for $\alpha = 0.00001$.)

f	conductivity ratio $\alpha = 0.1$		conductivity ratio $\alpha = 0.00001$	
	$C = 0.2$	$C = 1$	$C = 0.2$	$C = 1000$
0.000	1.00000	1.00000	1.00000	1.00000
0.050	0.97030	1.04079	0.96296	1.15765
0.100	0.94118	1.08271	0.92683	1.33280
0.150	0.91262	1.12579	0.89156	1.52866
0.200	0.88461	1.17010	0.85713	1.74941
0.250	0.85711	1.21566	0.82348	2.00070
0.300	0.83012	1.26254	0.79058	2.29057
0.350	0.80360	1.31077	0.75836	2.63091
0.400	0.77750	1.36038	0.72678	3.04033
0.425	0.76461	1.38571	0.71120	3.27996
0.450	0.75180	1.41139	0.69575	3.55020
0.475	0.73908	1.43742	0.68041	3.85924
0.500	0.72644	1.46381	0.66517	4.21900
0.525	0.71387	1.49054	0.65002	4.64759
0.550	0.70136	1.51762	0.63494	5.17446
0.575	0.68889	1.54503	0.61990	5.85154
0.600	0.67646	1.57277	0.60487	6.78214
0.625	0.66404	1.60082	0.58981	8.21162
0.650	0.65160	1.62916	0.57464	10.9324
0.660	0.64661	1.64057	0.56851	13.0137
0.670	0.64161	1.65202	0.56234	16.7836
0.680	0.63659	1.66351	0.55608	26.4975

of 64 spherical inclusions, which is two times larger than that of Bonnecaze & Brady (1991). We did not try larger systems, because our calculations were done only on a work station. The results are convergence tested, i.e. we increase the number of multipole modes slowly for a typical configuration at specified volume fraction and conductivity ratio so that the solutions converge to the reported digit. Then we use this number of modes to calculate the results for the desired volume fraction and conductivity ratio.

Table 4 lists the 95% confidence intervals of the dimensionless effective conductivity for conductivity ratios of 0.1 and 0.00001, each with two different values of interfacial conductance parameter C. The random configurations are generated by a standard Monte Carlo simulation procedure. The statistical data is obtained using 35 realizations of the random distributions. Convergence difficulties limited us from obtaining data for volume fractions higher than 0.5. Figure 4 shows the results

Table 3. *Dimensionless effective thermal conductivity $\sigma_{\mathrm{eff}}/\sigma_1$ of face-centred cubic lattice of spherical inclusions with imperfect interfaces versus the volume fraction f*

(Here $f_{\mathrm{max}} = (\sqrt{3}/8)\pi$. The critical value $C_{\mathrm{c}} = 0.45$ for $\alpha = 0.1$ and $C_{\mathrm{c}} = 0.499\,995$ for $\alpha = 0.000\,01$.)

	conductivity ratio $\alpha = 0.1$		conductivity ratio $\alpha = 0.000\,01$	
f	$C = 0.2$	$C = 1$	$C = 0.2$	$C = 1000$
0.000	1.000 00	1.000 00	1.000 00	1.000 00
0.050	0.970 30	1.040 79	0.962 96	1.157 65
0.100	0.941 18	1.082 71	0.926 83	1.332 79
0.150	0.912 62	1.125 79	0.891 57	1.528 60
0.200	0.884 61	1.170 10	0.857 13	1.749 14
0.250	0.873 55	1.215 67	0.823 49	1.999 86
0.300	0.830 14	1.262 55	0.790 61	2.288 26
0.350	0.803 62	1.310 79	0.758 42	2.625 15
0.400	0.777 55	1.360 42	0.726 87	3.026 70
0.425	0.764 67	1.385 76	0.711 31	3.259 17
0.450	0.751 88	1.411 46	0.695 88	3.518 50
0.475	0.739 18	1.437 51	0.680 57	3.810 70
0.500	0.726 56	1.463 92	0.665 38	4.143 94
0.525	0.714 01	1.490 69	0.650 27	4.529 58
0.550	0.701 53	1.517 80	0.635 24	4.984 14
0.575	0.689 10	1.545 26	0.620 27	5.532 66
0.600	0.676 71	1.573 06	0.605 33	6.215 64
0.625	0.664 36	1.601 19	0.590 40	7.104 12
0.650	0.652 02	1.629 63	0.575 45	8.338 15
0.675	0.639 67	1.658 36	0.560 44	10.2466
0.700	0.627 29	1.687 37	0.545 30	13.8718
0.710	0.622 32	1.699 05	0.539 19	16.6124
0.720	0.617 34	1.710 76	0.533 03	21.4697
0.730	0.612 34	1.722 50	0.526 82	33.2305
0.740	0.607 32	1.734 28	0.520 54	123.366

for $\sigma_2/\sigma_1 = 0.1$, at interfacial conductance parameters of $C = 0.2$, $C_{\mathrm{c}} = 0.45$ and $C = 1.0$. We also included in this figure the tight bounds obtained by Torquato & Rintoul (1995) for the cases $C = 0.2$ and $C = 1.0$. For the parameters that we have considered here, it can be seen that the superconducting interface affects the effective conductivity in the same way as in the regular array case.

4. An accurate approximation formula

In this section, we will examine the accuracy of an approximation formula, (4.1), based on the three-point microstructural parameter ζ_2 for our interfacial supercon-
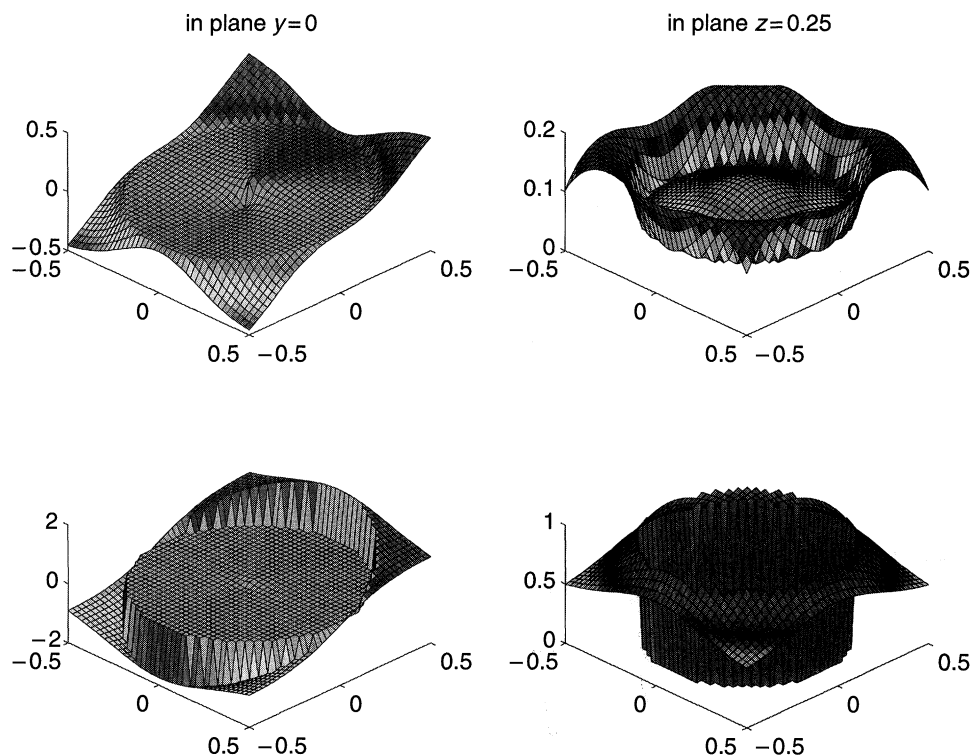
in plane $y = 0$ in plane $z = 0.25$

Figure 3. The surface plot of $\sigma(x)\partial T/\partial r$ in plane $y = 0$ (first column) and plane $z = 0.25$ (second column). The material is composed of a simple cubic lattice of spheres with conductivity ratio $\alpha = 0.1$ and volume fraction $f = 0.5$. The first row is for $C = 0$ (perfect interface), the second is for $C = 1$. The jump of the $\sigma(x)\partial T/\partial r$ at the interface is shown clearly in the latter case.

Table 4. *The 95% confidence intervals for the averaged dimensionless effective thermal conductivity* $\sigma_{\mathrm{eff}}/\sigma_1$ *of random suspensions of spherical inclusions with imperfect interfaces versus the volume fraction* f

(The critical value $C_c = 0.45$ for $\alpha = 0.1$ and $C_c = 0.499\,995$ for $\alpha = 0.000\,01$.)

	conductivity ratio $\alpha = 0.1$		conductivity ratio $\alpha = 0.000\,01$	
f	$C = 0.2$	$C = 1$	$C = 0.2$	$C = 1000$
0.1	0.9410 ± 0.0001	1.0831 ± 0.0002	0.9267 ± 0.0001	1.300 ± 0.003
0.2	0.8844 ± 0.0001	1.1708 ± 0.0004	0.8566 ± 0.0002	1.683 ± 0.006
0.3	0.8298 ± 0.0002	1.2635 ± 0.0005	0.7895 ± 0.0002	2.16 ± 0.01
0.4	0.7767 ± 0.0002	1.3616 ± 0.0005	0.7249 ± 0.0004	2.75 ± 0.02
0.5	0.7256 ± 0.0002	1.4650 ± 0.0007	0.6636 ± 0.0004	3.55 ± 0.03

ductance case. In the perfect interface case, such a formula was shown by Torquato (1985) to be able to predict the effective conductivity remarkably well for both periodic and random arrays of inclusions and for a wide range of volume fractions and conductivity ratios. We have shown in paper I that an appropriately generalized
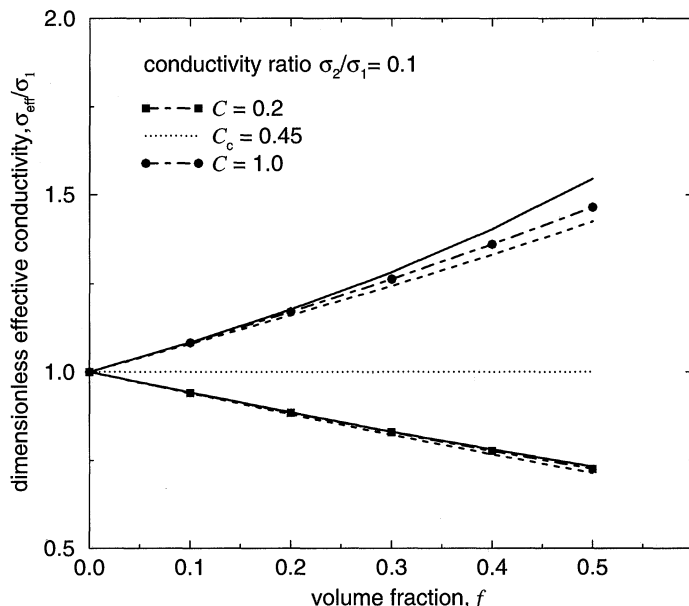
Figure 4. Dimensionless effective conductivity $\sigma_{\text{eff}}/\sigma_1$ versus volume fraction f for random case with sphere to matrix conductivity ratio $\alpha = \sigma_2/\sigma_1 = 0.1$. The filled circles indicate our calculated results for a moderate interfacial conductance $(C = 0.2)$ case, while the filled squares are for a large interfacial conductance $(C = 1)$ case. The dotted line is for the critical interfacial conductance value $(C_c = 0.45)$. The solid and dashed lines are the corresponding upper and lower bounds obtained from Torquato & Rintoul (1995) for each case.
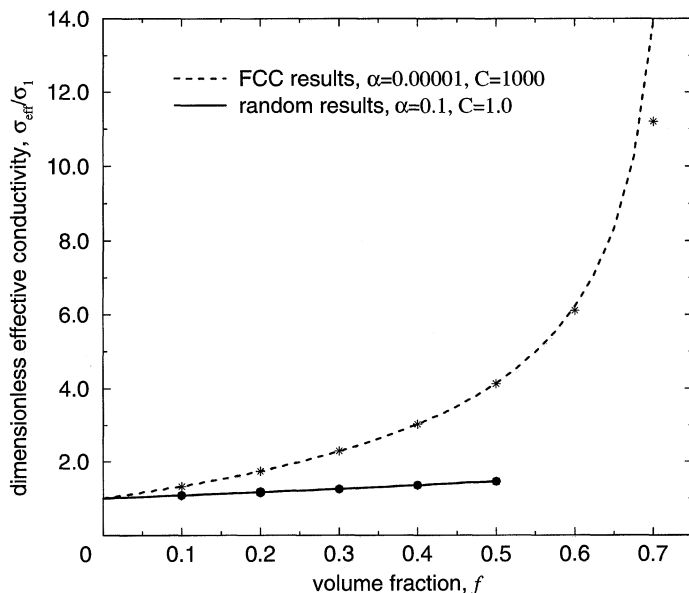

Figure 5. Comparison of the dimensionless effective conductivity $\sigma_{\text{eff}}/\sigma_1$ versus volume fraction f by formula (4.1) to exact calculations. The dashed line is the exact numerical result from § 3 for the face-centred cubic lattice case with conductivity ratio $\alpha = 0.00001$ and interfacial superconductance $C = 1000$, while the solid line is for the random dispersion with $\alpha = 0.1$ and $C = 1.0$. The stars and filled circles represent the respective predicted values by formula (4.1).

version of the formula for the interfacial resistance case, suggested by Torquato & Rintoul (1997), can also predict the effective conductivity remarkably well.

The approximate formula for the superconducting interface case given by Torquato & Rintoul (1997) is a trivial generalization of the one for the resistance case. The effective conductivity σ_{eff} of a suspension (random or periodic) with the dimensionless conductance C can be approximated by,

$$\frac{\sigma_{\text{eff}}}{\sigma_1} = \frac{1 + 2f\beta_1 - 2(1-f)\zeta_2\beta_1^2}{1 - f\beta_1 - 2(1-f)\zeta_2\beta_1^2}, \tag{4.1}$$

where the parameter β_1 depends on C via (2.16), and ζ_2 is the aforementioned three-point microstructural parameter. Once again, as in the perfect interface case, β_1 reduces to the parameter β_{21} contained in the approximate formula of Torquato (1985). For the three cubic lattices of spheres, ζ_2 has been tabulated as a function of the volume fraction f by McPhedran & Milton (1981). For random configurations, it was tabulated by Miller & Torquato (1990).

Figure 5 compares the exact numerical results for $\sigma_{\text{eff}}/\sigma_1$ from § 3 to corresponding results predicted by formula (4.1) for: (1) the face-centred cubic lattice case with conductivity ratio $\alpha = 0.00001$ and conductance parameter $C = 1000$ (dashed line); and (2) random suspensions with $\alpha = 0.1$ and $C = 1.0$ (solid line). The stars and filled circles represent the respective predicted values by formula (4.1). It is seen that formula (4.1) provides an excellent estimate of $\sigma_{\text{eff}}/\sigma_1$ for different values of conductivity ratio, interfacial conductance parameter and a wide range of volume fractions. Slight disagreement is observed only at the most extreme values of the parameters. The predictions of (4.1) for the simple and body-centred cubic lattice cases are equally good and hence are not shown graphically.

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