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Third-order model of thermal conductivity for random polydisperse particulate materials using well-resolved statistical descriptions from tomography

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ABSTRACT

For heterogeneous materials, obtaining an accurate statistical description has remained an outstanding problem. We accurately evaluate the three-point microstructural parameter that arises in third-order bounds and approximations of effective material properties. We propose new adaptive methods for computing n-point probability functions obtained from three-dimensional microstructures. We show that for highly packed systems our methods result in a 45% accuracy improvement compared to the latest techniques, and third-order approximations agree well with simulation data. Furthermore, third-order estimates of the effective behavior are computed for tomographically characterized systems of highly filled polydisperse ellipsoids and cuboids for the first time.

1. Introduction

The accurate characterization of many-body systems is a long studied problem with applications in many scientific fields at a variety of length scales from molecular arrangements [1,2] up to heterogeneous material microstructures [3] and celestial bodies [4–6]. Often, these systems have varying degrees of randomness at short and long range scales and often only lend themselves to statistical characterization. However, obtaining accurate higher order statistical correlations of many-body systems has proved difficult and is a limiting factor in understanding their physical processes.

Of particular interest in this Letter is determining effective transport and mechanical properties of many particle composites from higher order statistical data, which is a fundamental problem that has captured the attention of great minds including Einstein [7] and Maxwell [8]. The past fifty years have seen the formulation of rigorous bounds and approximations relying on a higher order statistical description of microstructures [9–12]. Especially third-order models have shown good agreement with experimental data [3]. While many theoretical advancements have been made for both linear and nonlinear material behavior [3,13,14], demonstration of these theories has been severely restricted to strong microstructural assumptions due to difficulties in accurately characterizing complex configurations. Various approximations have been formulated to describe complex configurations of random spheres [15–17], but direct computation of the statistical functions in three dimensions with resolution required in third-order models has been elusive.

In this Letter, we present adaptive methods rooted in computational mechanics and high performance computing for efficiently obtaining statistical functions without utilizing approximations of the shape and spatial configuration for random particulate systems. Third-order bounds and approximations of the effective thermal conductivity are computed using this high-fidelity statistical description. These computational methods, which we rigorously verify, allow for computation of statistical descriptors directly from complex three-dimensional microstructures with unprecedented accuracy. We show that previously formulated approximations of these statistical functions for systems of impenetrable monodisperse spheres result in significant inaccuracies, especially at higher volume fractions. Moreover, we extend these methods to other shapes, e.g. crystals, while accounting for degree of polydispersity. For the first time, we compute third-order bounds and approximations of the effective thermal conductivity for tomographically characterized three-dimensional packs of polydisperse ellipsoids and cuboids.

2. Theory of effective material behavior

In this Letter, we explore the effective behavior of steady-state heat conduction described by the conservation of energy assuming Fourier's law. Utilizing the variational principles of minimum

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energy and minimum complementary energy assuming ergodicity, statistical and material isotropy, Beran [18] derived third-order bounds of the effective conductivity, κ_e. Torquato [19] and Milton [10] independently simplified these bounds for two-phase composites to $\kappa^L \leq \kappa_e \leq \kappa^U$, where

$$\kappa^L = c_p\kappa_p + c_m\kappa_m - \frac{c_m c_p(\kappa_p-\kappa_m)^2}{c_m\kappa_p + c_p\kappa_m + 2\left(\frac{\zeta_p}{\kappa_p}+\frac{\zeta_m}{\kappa_m}\right)^{-1}}. \tag{1}$$

In this formulation, the bounds depend on the individual phase conductivities κ_i ($i = m$ (matrix) or p (particle)), the volume fractions c_i, and the microstructure parameters ζ_i. This microstructural parameter depends on the one-, two- and three-point probability functions ($S_i = c_i$, S_{ii}, and S_{iii}) and is defined as

$$\zeta_i = \frac{9}{2c_p c_m}\int \frac{3\cos^2\theta - 1}{2r_1 r_2}\tilde{S}_{iii}(r_1, r_2, \theta)\mathrm{d}(\cos\theta)\,\mathrm{d}r_1\,\mathrm{d}r_2, \tag{2}$$

where

$$\tilde{S}_{iii}(r_1, r_2, \theta) = S_{iii}(r_1, r_2, \theta) - \frac{S_{ii}(r_1)S_{ii}(r_2)}{c_i}. \tag{3}$$

For isotropic two-phase systems, Torquato [20] derived a three-point approximation (TPA) that has shown good agreement with simulations [21]. This estimate is given as

$$\frac{\kappa_e}{\kappa_m} = \frac{1 + 2c_p\beta_{pm} - 2c_m\zeta_p\beta_{pm}^2}{1 - c_p\beta_{pm} - 2c_m\zeta_p\beta_{pm}^2}, \tag{4}$$

where

$$\beta_{pm} = \frac{\kappa_p - \kappa_m}{\kappa_p + 2\kappa_m}. \tag{5}$$

While these bounds and approximations were derived decades ago, progress has been slow in accurately determining the microstructural parameter ζ_i (Eq. (2)) due to difficulties in computing the n-point probability function in Eq. (3). For highly filled random particulate composites, no one has been able to compute the probability functions from three-dimensional domains with the fidelity required by these third-order models. As analytical expressions of the n-point probability functions do not generally exist for composites with random configurations, a Monte Carlo-based statistical sampling algorithm is utilized. The accuracy of this method is $\mathcal{O}(1/\sqrt{N_r})$, where N_r is the number of random samples used to compute one function value of $S_{iii}(r_1, r_2, \theta)$, $S_{ii}(r)$, or S_i. A random sample consists of a random translation (described by 3 position values) and random rotation (described by three angles) of a $(n-1)$-simplex within the three-dimensional material domain. To speed up the analysis, we use high performance computing, where the N_r random samples are decomposed on $\mathcal{O}(10^3)$ computing cores. Others [22,23] have attempted a Monte Carlo sampling strategy to compute the probability functions, but on regular structured grids.

3. Adaptive interpolation and sampling method

Since computations on a structured grid are inefficient, we propose an adaptive triangulation technique. This method involves iteratively constructing a Delaunay triangulation of tetrahedrons, which we will refer to as $\mathcal{T}$, to create an interpolant of $\tilde{S}_{iii}(r_1, r_2, \theta)$ with C^0 continuity. Summarizing the algorithm, an initial regular tetrahedral grid is constructed for the domain $[r_1 = 0, r_1 = r^\infty] \times [r_2 = 0, r_2 = r_1] \times [\theta = 0, \theta = \pi]$ (note that this is half of the integration domain since the function is symmetric about the axis $r_1 = r_2$). This triangulation and associated

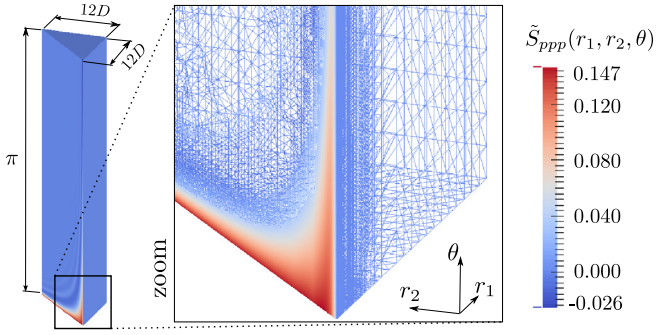

Fig. 1. Illustration of resulting adaptive triangulation of $\tilde{S}_{iii}(r_1, r_2, \theta)$ for system of impenetrable monodisperse spheres with $c_p = 0.6$.

function values, $\tilde{S}_{iii}(r_1, r_2, \theta)$ which appear in the integral kernel of Eq. (2), define the initial interpolant $\mathcal{T}_{l=0}$ (l is the adaptive iteration level). For a given iteration, a bisection method is used to refine the triangulation based on the local error of the statistics. For all tetrahedrons in a given iteration of the interpolant $\mathcal{T}_l$, the local accuracy is evaluated at each edge midpoint by considering the difference between the interpolated and computed probability functions. If an edge midpoint does not satisfy the error indicator function $\epsilon_a = \left|\tilde{S}_{iii}(r_1, r_2, \theta) - \mathcal{T}_l(r_1, r_2, \theta)\right| < tol$, where tol is a set tolerance, each edge of the tetrahedron is bisected and added to $\mathcal{T}_{l+1}$. If all midpoints in a tetrahedron satisfy ϵ_a, the tetrahedron is added to $\mathcal{T}_{l+1}$ unchanged. This iterative process is repeated until all grid points satisfy the error indicator function. After a convergence study, it was determined that $tol = (1/200)\max\{\tilde{S}_{iii}(r_1, r_2, \theta)\}$ results in low numerical error for all computations presented in this Letter. An example of the resulting triangulation for a highly filled ($c_p = 0.6$) monodisperse system of spheres with diameter D is shown in Fig. 1. Note that this function is 0 in a majority of the domain, but sharp variations exist near the origin ($r_1 = 0, r_2 = 0, \theta = 0$), along the diagonal $r_1 = r_2$ and near the edge of the domain where $r_1 = r_2 = 0$. The triangulation for this example contains $\mathcal{O}(10^7)$ tetrahedrons with a minimum edge length of $2.78 \cdot 10^{-17}D$, a mean edge length of $3.61 \cdot 10^{-2}D$, and a maximum edge length of $1.55D$, thus revealing the wide range of length scales required for accurately representing this function. The resulting interpolant $\mathcal{T}$ is then used as the basis for computing ζ_i via simplex integration of Eq. (2). In this work, Monte Carlo integration of each tetrahedron is performed. A convergence study determined that using $N_{int} = 1000$ random integration points per tetrahedron is sufficient for all microstructures presented in this Letter. Given that there are $\mathcal{O}(10^7)$ tetrahedrons in a typical interpolant, $\mathcal{O}(10^{10})$ integration points are required to evaluate the integral (2).

4. Verification

The proposed adaptive triangulation technique is verified by considering a two-phase system of overlapping spheres, which is one of a few configurations where the n-point probability functions of the matrix phase m can be defined analytically as:

$$S_{m\ldots m}(\boldsymbol{x}_1, \boldsymbol{x}_2, \ldots, \boldsymbol{x}_n) = \exp(-\rho V_n) \tag{6}$$

Here, ρ is the number density of spheres and V_n is the union volume of n spheres with diameter D. Considering four volume fractions of this model, ζ_m was computed with $r^\infty = 6D$. The results and errors associated with the computations are provided in Table 1. An error measure is defined as $\varepsilon_{PS} = \left|\zeta_m - \zeta_m^{R1}\right|/\zeta_m^{R1} \times 100$ [%], where ζ_m is the result from this work, and ζ_m^{R1} is the most accurate result presented in the literature [24]. Note that all