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Model and simulation predictions of the thermal conductivity of compact random nanoparticle composites

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ABSTRACT

The lattice thermal conductivity of compact random nanoparticle composites was investigated numerically as well as theoretically. By taking advantage of the three-dimensional Vorogoni diagrams, we were able to generate realistic numerical models of the composites. The phonon motion was then simulated in use of a Monte-Carlo simulator which employed an unstructured grid system (tetrahedron cells) and was developed under the single relaxation time approximation and the grey medium approximation. On the other hand, a new effective medium approximation (EMA) model was proposed in the present work to predict the thermal conductivity of nanoparticle composites which particles are all made of a same material type. This EMA model was next combined with the EMA model proposed by Liang and Ji (2000) for three bond percolation systems to predict the thermal conductivity of nanoparticle composites of two material types. It was found that with an effective particle diameter characterizing either the average particle volume or the average particle surface area, the model predictions agree excellently with the Monte-Carlo simulation results and match well with the measurements of nanostructured bulk alloys.

1. Introduction

Following the advance of nanotechnology, nanostructured materials have been designed and developed in the laboratories often for the sake of controlling the transport properties [1]. The embedded interfaces induce not only scatterings but also energy gaps to hinder low-energy carriers from passing. Favorable transport properties can be possibly obtained by manipulating the fabrication parameters. Among all, nanocomposites have received great attention recently because their fabrications are usually cheaper and their scale up is easier. Nanocomposites may be alloys fabricated by doping particular atoms in the crystal lattices (Brown et al. [2]), compact particle composites formed by hot pressing nanoparticles (Tang et al. [3], Mi et al. [4], Paul et al. [5]), or polycrystals condensed from solutions with special formula and cooling rates (Sootsman et al. [6]). Regardless of the fabrication methods, the compactness and thermal stability of the nanocomposites are the key issues that affect the transport properties. And any thermal instability leads to a merger and growth of the nanograins and consequently a loss of the nanostructures. In this work, we are particularly interested in exploring the influence of the nanostructures on the thermal conductivity of the composites.

It is well known that the interface density dominates the thermal transport property. An analytical relation between the interface density and the effective thermal conductivity however is still not found. Besides, both the particle geometry and the particle size distribution play important roles as well but have not been well explored. Most previous numerical studies treated nanocomposites with more or less regularity. Yang and Chen [7] and Huang and Kang [8] investigated the thermal conductivity of a matrix material embedded with regularly aligned nanowires. Liang and Ji [9] simulated the thermal transport in the composites of regularly stacked bulk cubes of random type of materials. In their study, the interfaces between the same type of cubes were removed to form clusters. Tien and Yang [10] replaced bulk cubes by square nanowires and studied the thermal conducitivity of the nanowire composites in use of Monte Carlo simulations. In all the above composites, continuous heat paths and consequently the thermal percolation may exist when the concentration of one of the constituents is large enough. The composites studied by Tian and Yang [11] are nanowires of two material types alternatively aligned to form a chess-board-like cross section. Hao et al. [12,13] performed a full-spectrum simulation and obtained the thermal conductivities of silicon nanoparticle composites with or without subparticles.

In theoretical analysis, there are two major ways of analyzing the disordered features of composites. The first one adopts the Callaway model [14,15]. However Callaway model needs a lot of experimental data for parameter fitting and is not convenient to

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be utilized. The second way employs the concept of the effective medium approximation (EMA). An EMA model treats a composite as a brand-new homogeneous material and looks for an effective transport property by using macro properties of its ingredients. It starts from the work of Landauer [15] for an estimate of the electric conductivity for two bond percolation systems. By adjusting the number of bonds in problem, Kirkpatrick [16] generalized the model to two dimensional (2D) square nets and three dimensional (3D) cubic nets of electric conductances. Liang and Ji [17] further extended this model to three bond percolation systems and used an averaged bond number to study the layer thickness effect on the thermal conductivity of disordered composite thin films. A satisfactory agreement was obtained. Phelan and Niemann [18] did a similar work by approximating the thin random composite material by a 2D network of thermal resistances. The thermal contact conductance between the filler and the matrix was included. Hasselman and Johnson [19] solved the diffusion equation and enforced the continuity conditions at the interface between a particle and the host material to obtain the effective thermal conductivity of particle-embedded-in-host composites under the assumption of a dilute particle concentration. The shape of particles could be spherical, columnar, or flat plate. Although Hasselman and Johnson's EMA model works pretty well on microcomposites while compared to experimental data, the dilute-particle-concentration assumption limits its application. When the composites are at nanoscale, Minnich and Chen [20] suggested a use of the Casimir limit to correct the phonon mean free paths inside the nanoparticles as well as inside the host material. A modified formula for spherical nanoparticle was thus derived based on the interface thermal resistance proposed by Chen [21] and more formulas were successively developed for various shapes of the nanoparticles [22,23]. While the interface thermal resistance arises from the interface scattering, so is the Casimir limit. When both are taken into consideration, the interface scattering effect is seemingly doubly counted. Besides, in all the above mentioned EMA models, the dilute particle concentration restriction does not get relaxed. Poon and Limtragool [24] ever attempted to propose an EMA model for arbitrary particle concentrations. Problem arises in their argument however as the concentration is close to one.

In this work, we aimed at obtaining a complete EMA model by following the philosophy of Poon and Limtragool's [24] but adopting a new translation of the composites to extend its application to particle-embedded-in-host composites with any arbitrary particle concentration, including 100%. Compact nanoparticle composites of a single material type are equivalent to particle-embedded-in-host composites with a 100% particle concentration. By combing our model with the EMA model proposed by Liang and Ji [17] for three bond percolation systems, we also targeted at predicting the thermal conductivity of compact nanoparticle composites of two material types. To confirm the reliability/accuracy of the proposed models, we attempted to compare the model predictions with the Monte-Carlo simulation results. In the simulations, we employed 3D Voronoi diagrams to mimic real nanoparticles; in other words, the shapes of the nanoparticles are irregular polyhedrons. Due to the irregularity, the Monte-Carlo simulator was developed based on 3D unstructured grids formed by tetrahedron cells. As before both the single relaxation time approximation and the gray medium approximation were employed for reducing the computational amount [25]. A comparison was also made with the full-spectrum simulations [12,13] in order to examine the validity of the grey medium approximation.

The rest of the paper is arranged as follows. The existing and newly proposed EMA models are introduced in Section 2. The fundamentals and verification of the unstructured-grid-based Monte Carlo simulator are described in Section 3. The results and discussions are presented in Section 4. In Section 5 are the conclusions.

2. EMA models for nanocomposites

2.1. Hasselman and Johnson's EMA model

The EMA model proposed by Hasselman and Johnson [19] for the particle-embedded-in-host composites is first recalled as follows

$$\frac{k_{eff}(\phi)}{k_h} = \frac{k_p(1+2\alpha)+2k_h+2\phi[k_p(1-\alpha)-k_h]}{k_p(1+2\alpha)+2k_h-\phi[k_p(1-\alpha)-k_h]} \tag{1}$$

where appear the effective thermal conductivity k_{eff}, the particle concentration ϕ, the dimensionless interface thermal resistance $\alpha = 2R_0k_h/D$, and the bulk thermal conductivities of particle and host materials k_p and k_h; D is the diameter of the particles and R_0 is the dimensional interface thermal resistance between the host and the particle. According to Chen's suggestion [21], we use the equilibrium temperatures at both sides of the interface to define the interface thermal resistance and thus write

$$R_0 = \frac{2(C_h\upsilon_{g,h}+C_p\upsilon_{g,p})}{C_h\upsilon_{g,h}C_p\upsilon_{g,p}} \tag{2}$$

where C_h and C_p are the specific heats and $\upsilon_{g,h}$ and $\upsilon_{g,p}$ are the phonon group velocities of the host and particle materials, respectively.

When ϕ equals to one, the particle-embedded-in-host composite becomes a compact nanoparticle composite of a single material type. However, in deriving Eq. (1), Hasselman and Johnson assumed a dilute particle concentration, i.e. $\phi << 1$. Therefore it is not consistent to directly apply $\phi = 1$ in Eq. (1).

2.2. Arbitrary concentration model

Based on Hasselman and Johnson's work and following the philosophy of Poon and Limtragool [24], for a particle-embedded-in-host composite with an arbitrary particle concentration $\phi + d\phi$, we proposed a modified EMA model to predict its thermal conductivity as follows

$$\frac{k_{eff}(\phi+d\phi)}{k_{eff}(\phi')} = \frac{k_p(1+2\alpha')+2k_{eff}(\phi')+2d\phi[k_p(1-\alpha')-k_{eff}(\phi')]}{k_p(1+2\alpha')+2k_{eff}(\phi')-d\phi[k_p(1-\alpha')-k_{eff}(\phi')]} \tag{3}$$

where $\alpha' = \alpha(\phi') = 2R(\phi')k_{eff}(\phi')/D$. The idea behind the Eq. (3) is to translate the composite as a composite having some new host material and a new particle concentration $d\phi$. The new host material itself is a particle-embedded-in-host composite. To preserve the particles, the particle concentration ϕ' of this new host material must be $\phi' = \phi/(1-d\phi)$. Hasselman and Johnson's formula is applicable to this newly translated composite now because the new particle concentration $d\phi$ is infinitesimal. Therefore we replaced k_h by $k_{eff}(\phi')$ and R_0 by $R(\phi')$, the interface thermal resistance between the new host material and the particle. By taking the Taylor series expansion of Eq. (3) and taking the limit $d\phi \to 0$, we obtained

$$\frac{dk_{eff}}{k_{eff}} = \frac{3d\phi}{1-\phi}\left\{\frac{k_p(1-\alpha(\phi))-k_{eff}}{k_p(1+2\alpha(\phi))+2k_{eff}}\right\} \tag{4}$$

where $\alpha(\phi) = 2R(\phi)k_{eff}(\phi)/D$. With the initial condition $k_{eff}(0) = k_h$, the effective thermal conductivity of the particle-embedded-in-host composites with an arbitrary concentration ϕ is obtainable by integrating Eq. (4) once the interface thermal resistance $R(\phi)$ is known [24].

It must be mentioned that Eq. (4) is actually as the same as the formula derived by Poon and Limtragool [24] without the modifications of the phonon mean free paths within the particles and the host according to the Casimir limit. Poon and Limtragool however translated the interested composite as a combination of a composite host (having a particle concentration ϕ) and a new par-