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Simple model for effective thermal conductivity of bulk nanostructured materials

ChangJin Choi, Nicholas Roberts*

Department of Mechanical and Aerospace Engineering, Utah State University, Logan, UT 84322-4130, USA

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ABSTRACT

The ability to design and predict effective thermal transport properties in bulk nanostructured materials is becoming increasingly important as the demand for higher performance and higher efficiency electronics and energy conversion devices grows. A tremendous effort has been focused on understanding and improving our ability to predict thermal transport at an individual interface, resulting in new models that account for phonon populations and electron–phonon coupling. Despite the success of this work, there has been no effort to produce a simple analytic model to predict the effective thermal conductivity of bulk materials that include nanoscale features.

This paper focuses on applying knowledge of thermal interface resistances to the development of a simple analytic model that can be used for the efficient prediction of effective thermal transport behavior to aid in the design of thermoelectric materials and microelectronic devices. To this end, molecular dynamics simulations are performed to generate an initial model based on the effective thermal conductivity of in-plane and cross-plane superlattices and embedded nanoparticle and nanowire arrays. The model is then validated and generalized by comparing to existing computational and experimental data. Results shows that the effective thermal conductivity calculated from the analytic model agrees well with that of various systems of different materials and geometries, and provides the ability to predict effective system thermal conductivity of these material systems with variable interface area for optimization of thermal transport properties.

1. Introduction

The ability to predict thermal conductivity of bulk nanostructured materials (with the inclusion of nanparticles, nanowires, interfaces, and core–shell structures) is becoming increasingly important [1–4]. Over the past several decades, extensive research has been focused on understanding thermal transport behavior (electrons and phonons) across individual nanostructures, nanofeatures, or interfaces [5]. These studies have led to an improved fundamental understanding of thermal transport, but have not made the same advancement towards the ability to predict an effective thermal conductivity due to the inclusion of these features. Nan et al. showed, using an effective medium approximation, that the effective thermal conductivity can be significantly altered by the inclusion of interfacial thermal boundary resistance and is dependent on particle shape and size [6]. The effective medium approximation was more recently modified to account for the particle interaction at higher volume fraction to predict the effective thermal conductivity of nanocomposites [7]. However, these studies do not take into account the effect of thermal boundary resistance with respect to the specific interface orientations.

Ideally, a predictive model of this sort would include the multiscale physics that are present in these materials, but the only reasonable approach requires solution of the Boltzmann Transport Equation (or other computationally intensive approach), which is not realistic due to the intensive computational requirements of the solution and the necessary knowledge of the dispersion relation of the energy carriers [8]. This work proposes a simple analytic model for the calculation of an effective thermal conductivity of a bulk nanostructured material by considering the thermal boundary resistance associated with interfaces perpendicular and parallel to the direction of transport. The required information necessary for accurate use of this model is the thermal boundary resistance (or impact of a single interface on the thermal conductivity) of a single interface at each orientation and an estimate of the alloy limit

* Corresponding author.
E-mail address: nick.roberts@usu.edu (N. Roberts).

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Nomenclature

k	thermal conductivity
k_b	Boltzmann's constant
k_{AL}	alloy limit
k_{eff}	effective thermal conductivity
k_{Ge}	thermal conductivity of germanium
k_{Si}	thermal conductivity of silicon
n_{Si}	atomic percentage of silicon
q	heat flux
A	cross-sectional area
$A_{\parallel}$	interface area parallel to the heat flow
$A_{\perp}$	interface area perpendicular to the heat flow
T	temperature
ΔT	temperature difference
K	kinetic energy
L	length
N	number of atoms
ξ	thermal boundary resistance caused by parallel interface area
Ξ	thermal boundary resistance caused by perpendicular interface area

thermal conductivity of the material system. The limitations of this model are that (1) it only applies when wave-effects are not expected to be present (when nanoscale features are greater than the length scales associated with coherent phonon transport) [9–11] and (2) for systems in which electron contributions to thermal transport can be neglected, (i.e. metals and metal alloys).

The availability of a simple model, like the one presented here, could have a dramatic impact on the development of higher efficiency thermoelectric materials [12,13] allowing for the direct harvesting of a large amount of energy that is typically lost in the form of waste heat [14]. This model would also allow for the continued development of high performance electronic devices, where the thermal conductivity of the material or system could be predicted in advance, saving time, effort, and expenses associated with the "build and test" approach that is commonly applied. The alternatives to this simple model require detailed computational simulation that can also take considerable time and effort. These computational models have a distinct advantage in modeling the atomic and nanoscale physics, but fall well short of being able to model a full device or material [15]. This model takes advantage of the results obtained from simulations, like molecular dynamics (a powerful technique for studying transport behavior across a single or a few nanoscale features), for the prediction of an effective thermal conductivity of the material or system. This model can also take advantage of experimental measurements of individual nanoscale features that have been enabled through techniques like $3-\omega$, Raman spectroscopy, photothermal radiometry, thermal bridge, and the more recent time-domain and frequency-domain thermoreflectance techniques [16–22].

In this paper, an analytic model is developed from a set of molecular dynamics simulations and presented. The model accounts for the contribution of both perpendicular and parallel interface area on the effective thermal conductivity of a bulk nanostructured material using the popular Si/Ge material system. Many studies have been performed on the Si/Ge material system [23,24], thus sufficient data is available to validate our analytic model. Additionally, Si/Ge is a commonly used material system for microelectronics and high temperature thermoelectrics. The model is then applied to other material systems using computational and experimental results from the literature. This model provides the ability to predict an effective thermal conductivity with only a limited set of data from atomistic simulations or experimental measurements therefore saving time and money for the design of thermoelectrics and electronic devices.

2. Molecular dynamics method

Molecular dynamics (MD) is a common technique used to investigate thermal transport across individual nanoscale features [25–32]. Non-equilibrium MD (NEMD) is especially useful in the study of thermal boundary conductance across interfaces between similar and dissimilar materials. These simulations provide n × 8 × 8 both information about the thermal boundary conductance associated with the interface and the effective thermal conductivity of the material in which the interface exists. A review of NEMD and the other commonly used approach of molecular dynamics for determination of thermal transport properties (equilibrium MD) can be found in Ref. [33]. In this study NEMD simulations of Si and Ge systems are performed using the Stillinger–Weber interatomic potential [34–37] to formulate a model for effective thermal conductivity of bulk nanostructured materials. The NEMD simulations are all performed using LAMMPS [38].

The first set of materials used in this study are thin film structures constructed from bulk Si and Ge. The size of these materials are varied in length with a size of unit cells (UCs), where n = 32, 64, 128, and 256, with an additional 16 UCs (8 on each side) in the direction of heat flow for fixed position walls and heat source and sink. 4 UCs (2 on each side) are assigned to the fixed wall to ensure no energy transport directly from the heat source to the heat sink, and 12 UCs (6 on each side) are used as heat source and sink. The second set of materials are Si–Ge superlattice structures where the stacking sequences are parallel (in-plane) and perpendicular (cross-plane) to the heat flow direction. The third set of materials are embedded Ge–Si nanowire arrays and embedded Ge–Si nanoparticle arrays, as they are shown in Fig. 1.

A time step of 0.5 fs is used for all NEMD simulations. The zero pressure NPT (constant number of atoms, pressure, and temperature) ensemble is performed for a minimum of 30 ps under the periodic boundary condition in order to relax the lattice. In the main NEMD simulation, periodic boundary conditions are used in

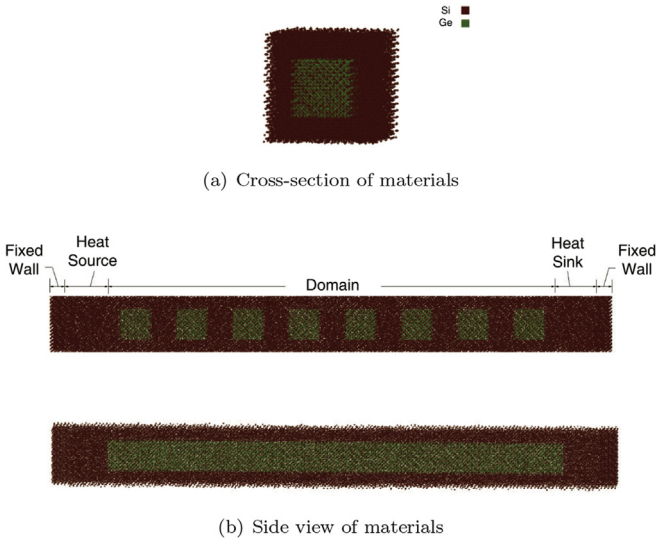

Fig. 1. Core-shell wire array (a) Interfacial area of the Ge–Si core–shell structure, (b) Embedded nanoparticle array (top) Embedded NW array (bottom).