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Phonon Boltzmann Transport Equation based modeling of time domain thermo-reflectance experiments

Syed Ashraf Ali, Sandip Mazumder *

Department of Mechanical and Aerospace Engineering, The Ohio State University, Columbus, OH 43210, USA

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ABSTRACT

Time Domain Thermo-Reflectance (TDTR) experiments have been recently identified as a viable pathway toward extracting the phonon mean free path spectrum of semiconductor materials. However, this requires an intervening model. It is now widely believed that the frequency and polarization dependent phonon Boltzmann Transport Equation (BTE) is the most suitable model for this purpose. In this article, TDTR experiments are simulated using large-scale parallel computations of the phonon BTE in a two-dimensional computational domain. Silicon is used as the candidate substrate material. Simulations are performed for multiple pulse and modulation cycles of the TDTR pump laser. This requires resolution of a picosecond laser pulse within a computational timeframe that spans several hundreds of nanoseconds. The metallic transducer layer on top of the substrate is modeled using the Fourier law and coupled to the BTE within the silicon substrate. Studies are conducted for four different laser spot sizes and two different modulation frequencies. The BTE results are fitted to the Fourier law, and effective thermal conductivities are extracted. It is demonstrated that the time delay of the probe laser could have a significant impact on the fitted (extracted) thermal conductivity value. The modulation frequency is found to have negligible effect on the thermal conductivity, while the spot size variation exhibits significant impact. Both trends are found to be in agreement with experimental observations. The thermal conductivity accumulation function is also computed, and the effect of the mean free path spectrum on the thermal conductivity suppression is delineated.

1. Introduction

Heat conduction in semiconductor materials primarily occurs by vibrational waves traveling through the lattice. From a quantum mechanical standpoint, these waves may be thought of as discrete energy packets, called phonons [1–3]. The mean free path of phonons in silicon at room temperature is approximately in the range 10–10^5 nm, with the average around 300 nm [3], which could be significantly larger than the characteristic feature sizes in modern-day applications of semiconductor materials. If the mean free path of the phonons is larger than or of the same order of magnitude as the characteristic length scale, thermodynamic equilibrium ceases to exist, and the Fourier law of heat conduction, which assumes local equilibrium, becomes invalid. The Boltzmann Transport Equation (BTE) for phonons serves as a viable alternative [1,2,4] for the prediction of non-equilibrium heat conduction in semiconductor materials at the nanoscale.

One of the critical inputs to the phonon BTE is the phonon relaxation time-scale. In a pure semiconductor, since phonons relax to equilibrium primarily due to phonon-phonon interactions or intrinsic scattering, the scattering time-scale may be deemed the most important input to the BTE. Another important input is the wavevector-frequency or dispersion relationship. The slope of the dispersion curve provides the group velocity of the phonons. The mean free path of a phonon is the product of its group velocity and its scattering time-scale. Since the group velocity and scattering time-scale are both frequency dependent, phonons of different frequency have different mean free paths. The frequency dependent mean free path is often referred to as the mean free path spectrum. In a nanoscale system, not all phonons contribute equally to thermal transport. Phonons with large mean free paths (ballistic phonons) interact more frequently with geometric boundaries than with other phonons, while phonons with small mean free paths (diffusive phonons) engage in frequent inter-phonon scattering. Therefore, from an engineering standpoint, it is conceivable

* Corresponding author at: Department of Mechanical and Aerospace Engineering, The Ohio State University, Suite E410, Scott Laboratory, 201 West 19th Avenue, Columbus, OH 43210, USA.
E-mail address: mazumder.2@osu.edu (S. Mazumder).

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Nomenclature

c	specific heat capacity per unit volume [J m^{-3} K^{-1}]
D	density of states per unit volume [m^{-3}]
f	number density function
f_0	equilibrium number density function
$G_{\omega,p}$	spectral directionally integrated intensity [W m^{-2} - rad^{-1} s]
$\hbar$	Dirac (or reduced Planck) constant = 1.0546×10^{-34} [m^2 kg.s^{-1}]
$I_{\omega,p}$	spectral directional phonon intensity [W m^{-2} sr^{-1} - rad^{-1} s]
$I_{0,\omega,p}$	equilibrium phonon intensity [W m^{-2} sr^{-1} rad^{-1} s]
k_B	Boltzmann constant = 1.381×10^{-23} [m^2 kg.s^{-2} K^{-1}]
$\hat{\mathbf{n}}$	unit surface normal vector
N_{band}	total number of spectral intervals (or bands)
N_{cell}	number of control volumes (or cells)
N_{dir}	number of control angles (or directions)
p	phonon polarization index
$\mathbf{q}$	heat flux vector [W m^{-2}]
$\dot{Q}$	heat transfer rate [W]
$\mathbf{r}$	position vector [m]
R_C	contact resistance [K W^{-1}]
$\hat{\mathbf{s}}$	unit direction vector
t	time [s]
T	absolute temperature [K]
u	internal energy per unit volume [J m^{-3}]
V_k	volume of cell k [m^3]

Greek

α	degree of specularity
θ	polar angle [radians]
κ	thermal conductivity [W m^{-1} K^{-1}]
Λ	mean free path [m]
$\boldsymbol{\upsilon}_{\omega,p}$	phonon group velocity vector [m s^{-1}]
$\tau_{\omega,p}$	spectral relaxation time scale [s]
ω	angular frequency [rad s^{-1}]
Ω	solid angle (sr)
ψ	azimuthal angle [rad]

that if the mean free path spectrum of phonons in a material was known, one could tailor the geometric structure in a manner that would either enhance or suppress the thermal conductivity of the nanostructure by allowing or disallowing phonons of certain frequencies to participate in thermal transport. Hence, determination of the phonon mean free path spectrum in semiconductor materials is currently considered a translational area of research within the nanoscale heat transfer community [5].

Of the aforementioned two quantities that constitute the mean free path, namely the dispersion relationship and the scattering time-scale, the scattering time-scale is the more challenging to determine. Computation of the scattering time-scale of various phonon modes and frequencies is an active area of research. Starting from the works of Callaway [5] and Holland [6,7] in the 1960s, various approaches have been used to estimate three-phonon scattering rates. These include perturbation techniques pioneered by Klemens [8] in combination with the selection rules of collision, molecular dynamics [10–14], collision integral methods [15–18], *ab initio* methods combined with Fermi's golden rule [19–24], among others. As to which method is most appropriate for the task at hand is a highly controversial subject. Worth noting is the variability in the values of the scattering time-scales predicted by various methods [8,9,14,18,20]. Despite significant variability in the predicted scattering time-scales, researchers in this area typically demonstrate "good" agreement between their predicted effective thermal conductivity and some selected experimental data. Although the common notion is that the scattering time-scales predicted by first-principles methods [19–24] are more accurate than the time-scales proposed by earlier works [8,9], there is no direct evidence in the form of a comparative study to support this notion. More importantly, thermal conductivity is the net manifestation of the propagation of phonons of many different modes and frequencies and their interactions with geometric boundaries, and agreement (model versus experiment) in effective thermal conductivity neither validates nor disproves the predicted scattering time-scales. What is needed is direct validation of the mean free path spectrum against experimental measurements, or conversely, its direct extraction from experimental data, *i.e.*, without taking the thermal conductivity route.

Experiments at the nanoscale are limiting in scope. Until recently, experiments have only been able to provide effective thermal conductivity of thin semiconductor films as a function of film thickness, temperature, and dopant concentrations [4]. While it is possible now to measure phonon relaxation times directly using inelastic neutron scattering, X-ray scattering, or Brillouin scattering [25,26], the data obtained by such methods do not span the entire frequency range of the phonons that are believed to be the carriers of heat [4,27]. In the past decade, a new class of experiments—so-called laser based pump probe experiments—hold the promise of divulging significantly more information than just the effective thermal conductivity of the material under scrutiny [27–33]. Experimental techniques that belong to this category are transient thermal grating, time domain thermo-reflectance (TDTR), frequency domain thermo-reflectance, and their variations. An excellent recent review of these techniques is provided by Regner et al. [5]. Although the details are somewhat different in these experiments, they all share the common philosophy of heating a designated spot on a substrate with one laser beam (the pump laser) and monitoring temperature evolution with another one (the probe laser). The temperature response of the hot spot is finally analyzed (fitted to a model) to extract the thermal conductivity. Furthermore, in approaches based on thermo-reflectance, the detection principle requires the power of the heating laser to be modulated, and in the case of TDTR, a pulse train is implemented, thereby enabling—at least, in principle—identification of the dominant phonon frequencies that contribute to thermal transport and the effective thermal conductivity.

It is now unanimously acknowledged [5] that the BTE for phonons is capable of describing non-equilibrium thermal transport in semiconductors, and is an appropriate intervening model for describing the causal relationship between the spectral scattering time-scale of phonons, the geometry in question, and the resulting temperature and heat flux distributions. The determination of the spectral scattering time-scale from measured temperature, of course, constitutes an inverse problem. Nonetheless, prior to solution of this inverse problem, it is first necessary to conduct forward calculations that will predict temperature and heat flux with the spectral scattering time-scale as inputs. This implies that if TDTR is the experimental technique being used, one must first be able to simulate TDTR experiments exactly using the modeling framework, namely the multi-dimensional frequency-dependent phonon BTE. This is a critical barrier that must first be overcome [5]