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Thermal Transport Modeling Of Semiconductor Materials From First Principles

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THERMAL TRANSPORT MODELING OF SEMICONDUCTOR MATERIALS FROM FIRST PRINCIPLES

A Thesis Presented

by

ALIYA QURESHI

Submitted to the Graduate School of the University of Massachusetts Amherst in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE IN ELECTRICAL AND COMPUTER ENGINEERING

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Electrical and Computer Engineering
THERMAL TRANSPORT MODELING OF SEMICONDUCTOR MATERIALS FROM
FIRST PRINCIPLES

A Thesis Presented
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To my mother.
ACKNOWLEDGMENTS

I would like to express my deepest appreciation to my advisor, Professor Zlatan Aksamija, you have been a tremendous mentor for me. I would like to thank you for your thoughtful patience, guidance and support throughout and allowing me to grow. A special thanks to Professor Neal Anderson and Professor Eric Polizzi for kindly serving as my thesis committee.

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ABSTRACT

THERMAL TRANSPORT MODELING OF SEMICONDUCTOR MATERIALS FROM FIRST PRINCIPLES

MAY 2020

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Over the past few years, the size of semiconductor devices has been shrinking whereas the density of transistors has exponentially increased. Thus, thermal management has become a serious concern as device performance and reliability is greatly affected by heat. An understanding of thermal transport properties at device level along with predictive modelling can lead us to design of new systems and materials tailored according to the thermal conductivity. In our work we first review different models used to calculate thermal conductivity and examine their accuracy using the experimentally measured thermal conductivity for Si. Our results suggest that empirically calculated rates used in thermal conductivity calculations do not capture the scaling behavior for three phonon scattering mechanism properly. This directly affects the estimation of the thermal conductivity and therefore we need to capture them more accurately. Also, we observe that at low temperature the Callaway and the improved Callaway model show good agreement where boundary scattering is dominant, whereas at high temperature iterative and RTA models show good agreement where three-phonon scattering is dominant. Therefore, their
lies a need for a model which can characterize K properly at low and high temperature. Second, we then calculate the three phonon scattering rates using first-principles and combine them into the Callaway model. Through our work we successfully build a hybrid model which can be used to describe thermal conductivity of Si for a temperature range of 10 K to 425 K which captures the thermal conductivity accurately. We also show that in case of Si the improved Callaway model and Callaway model both perform equally well.
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CHAPTER 1
INTRODUCTION

Device engineers have been ingenious at maintaining Moore’s Law, with shrinking the size and escalating density of transistors and other integrated circuit devices. Modern Si microelectronics are now firmly in the nanoscale regime with enhanced computing capabilities, but at the cost of increasing power dissipation across the device and system levels. Thermal management is a serious issue as the size of devices and structures decrease, as device performance is being significantly affected by temperature. Hence, understanding of the heat transport details are required which can lead to material designs with better performance. Theoretical models developed in parallel with experiments can offer great insights into the different mechanism which affect transport properties. A computational technique that has a predictive power will thus greatly facilitate in understanding of heat dissipation nanoelectronics and will also assist in the optimization of existing materials and the search for better ones to improve the overall performance of the working device [1].

The lattice thermal conductivity is a key property in countless applications. Important technologies that demand specific materials with tailored thermal conductivities include thermoelectricity, heat management [2]. Through the predictive modeling of thermal conduction, we can conceive a materials-by-design approach in which natural materials are manipulated at the nanoscale, resulting into new systems with heat transport properties tailored to meet technology-relevant issues and needs [3]. Heat conduction in most semiconductors is dominated by lattice vibrational waves. The basic energy quantum of
lattice vibrations is called a phonon. Phonons can be treated as both waves and particles. There is an increasing interest in the theoretical prediction of material properties in which phonon play an essential role and therefore, an understanding of phonon transport is required. Thermal transport can be studied using molecular dynamics simulation or lattice dynamic calculations. However, evaluating the lattice thermal conductivity using molecular dynamics method lacks predictive power due to empirical interatomic potentials involved. The first principles driven lattice dynamics calculations are routine and is used by researchers to study a variety of simple semiconductors.

In our work, we perform lattice dynamics calculations using force constants obtained from first principles driven density functional theory to calculate the phonon-phonon scattering rates and calculate the thermal conductivity using Callaway model.

1.1 The Thermodynamics Picture

Heat, charge and mass flux in a condensed matter system are globally referred to as transport phenomena, which includes many aspects such as thermoelectricity, thermal fluctuation, harmonic oscillation, and phonon thermal conductivity. Fourier law of heat conduction has been used to describe most of the engineering problems dealing with transport, exchange, and accumulation of thermal energy, according to which the thermal energy transfers diffusively if the length of the system is much longer than the particle mean free path. The relation between heat flux is related to the temperature gradient can be expressed by the Fourier law as, where $K$ is the thermal conductivity

$$Q = - K \nabla T \quad (1).$$
In nanostructures the applicability of Fourier law is questionable because the length scales associated with the energy carriers become comparable to or larger than the characteristic length of nanostructure. Boltzmann introduced the method by using mechanics and statistics to model the heat conduction, entropy, and other thermodynamic properties [4, 23, 25]. Using the Boltzmann transport equation (BTE), phonon transport can be predicted for each phonon mode, when the domain length scales are larger than the dominant phonon wavelength (typically 2-3 nm for silicon at room temperature), but comparable to the phonon mean free path $\Lambda = 300$ nm. Figure 1.1 provides a general guideline for the appropriate treatment of phonon transport in different nanostructure. The BTE is the most widely used approaches for calculating thermal conductivity and will be our cornerstone for discussion.

Figure 1.1: Regime map for phonon transport in ultra-thin silicon layers, adapted from [26]
1.2 Boltzmann Transport Equation

When a material is at equilibrium at a temperature $T$, the equilibrium phonon population $n_{qj}^0$ describes the number of phonons for each vibrational normal mode $\omega_{qj}$ with wavevector $q$ and branch index $j$ found at energy $\hbar \omega_{qj}$. Starting from this condition of equilibrium, if a gradient of temperature $\nabla T$ is applied, the phonon population leaves its equilibrium condition, reaching an out-of-equilibrium condition. A non-zero heat current $J_h$, collinear to the gradient of temperature, is thus generated and is written as

$$J_h = \frac{1}{V} \sum_q \sum_j \hbar \omega_{qj} v_{qj} n_{qj} \ldots (2)$$

where $V$ is the crystal volume, $\hbar$ reduced Planks constant, $v_{qj}$ is the group velocity, $n_{qj}$ non-equilibrium population due to $\nabla T$. This situation is conceptualized in Figure 1.2.

![Figure 1.2](image.png)

Figure 1.2: Conceptual scheme of heat flux generation. A thermal gradient is applied to a homogeneous dielectric crystalline material generating a heat flux, which is determined by the net variation of the phonon population within each volume element, due to an unbalanced phonon population between the hot and cold ends and to phonon–phonon scattering. Adapted from [3].
The evolution of $n_{qj}$ at any given time, when temperature gradient is applied across a material is the key quantity to be evaluated which is achieved by the introduction of Boltzmann Transport Equation (BTE). At steady state with no external forces, the BTE is a balance between changes in phonon distribution due to drift and scattering arising from allowed processes

\[
\frac{\partial n_{qj}}{\partial t} \bigg|_{\text{scatter}} = v_{qj} \frac{\partial n_{qj}}{\partial T} \nabla T \quad \ldots (4).
\]

The phonons in a material can scatter from other phonons, isotopes, defects, impurities, grain/system boundaries, etc. and $\frac{\partial n_{qj}}{\partial t}$ denoting the total change in phonon distribution due to all scattering mechanisms. Equating the heat flux from equation (1) and (2) we get

\[
-K \nabla T = \frac{1}{V} \sum_q \sum_j h\omega_{qj} v_{qj} n_{qj} \ldots (3)
\]

where $K$ is the thermal conductivity.

**1.3 Thesis Outline**

The remainder of thesis is organized as follows: Chapter 2 reviews the phonon interaction and discuss about the three-phonon interactions. It also throws some light on the existing models for thermal conductivity calculations and their drawbacks. Chapter 3 reviews the methodology for the thermal conductivity calculations. It also discusses the results for thermal conductivity of Si obtained using first principles and analytically calculated scattering rates. Chapter 4 reviews the thermal conductivity calculations for diamond using different models.
CHAPTER 2
PHONON INTERACTIONS

Phonon scattering including phonon-phonon, phonon-boundary, and phonon-defect (e.g., impurities, dislocations, or vacancies) scattering affects the energy transport properties of insulators and semiconductors. Boundary scattering is the dominant process at low temperatures even in bulk materials [27, 28] which can be expressed as

$$(\tau_{\text{boundary}})^{-1} = \frac{c_s}{L_0} \ldots (5)$$

where $c_s$ is the average speed of sound and $L_0$ is the characteristic length of sample. At low frequencies anharmonic and impurity scattering rates are negligible compared to the boundary scattering rate which is on the order of $v/L$ and thus finite.

The other source of phonon scattering is defects or impurities, which is dominant at intermediate temperatures smaller than the Debye temperature ($\Theta_D$), where there are not too many phonons present. It is expressed as

$$(\tau_{\text{impurity}})^{-1} = \frac{\Pi}{6} V_o \Gamma_{\text{imp}} \omega^2 D(\omega) \ldots (6)$$

where $V_o$ is the volume per atom, $\Gamma_{\text{imp}}$ is the impurity constant and $D(\omega)$ is the density of states. For low-frequency, long wavelength phonons, the impurity scattering rate is proportional to the $\omega^4$ fourth power of the phonon frequency. This is the well-known Rayleigh scattering limit. Finally, at higher temperatures, near and above the $\Theta_D$ where the phonon concentration is large, three or more phonon scattering processes dominate transport also called as anharmonic scattering. These phonon-phonon interactions come from the anharmonic terms of crystal potential energy expansion and anharmonicity itself is the result of significant deviation of atoms from their equilibrium position. These
scatterings are further divided into two categories \textit{normal} (N) process and \textit{umklapp} (U) process, which will be discussed in the sections followed. These are expressed

\[ (\tau_{\text{anharmonic}})^{-1} = (\tau_{\text{normal}})^{-1} + (\tau_{\text{umklapp}})^{-1} \]

\[ (\tau_{\text{umklapp}})^{-1} = B_{U} \omega^{3-4} T e^{-\theta/3T} \ldots (7) \]

\[ (\tau_{\text{normal}})^{-1} = B_{N} \omega^{a T} \ldots (8) \]

where \( B_{U} = \frac{\hbar v_{q}^{2}}{M v_{q}^{2} \theta} \), \( B_{N} = \left( \frac{k_{B}}{\hbar} \right)^{b} \frac{\hbar v_{q}^{2} V^{(a+b-2)/3}}{M v_{q}^{a+b}} \), \( M \) is average atomic mass of carbon, \( \bar{\gamma} \) is Gruneisen’s parameter, \( V \) is volume of unit cell \cite{35} . These analytical rates are approximated using experimental results of existing materials. However, these are not necessarily predictive for new materials and therefore the usage of first principles calculated rates.

Figure 2.1 shows the regimes of different scattering mechanisms in a conductivity vs. temperature plot. When all processes are present and compete, the combination rule that gives the total relaxation time, according to which the rates of different independent processes add to give the total scattering rate.

\[ \frac{1}{\tau_{\text{total}}} = \frac{1}{\tau_{\text{boundary}}} + \frac{1}{\tau_{\text{defect}}} + \frac{1}{\tau_{\text{anharmonic}}} \ldots (9) \]

### 2.1 Anharmonic Scattering

During scatterings, energy and momentum conservation need to be satisfied, which impose the conditions on which processes are allowed. The momentum conservation requires that the three-phonon wave vectors involved in the scattering add up to zero or up to an integer number of reciprocal lattice vectors. As shown in the Figure 2.2, phonon-
phonon scattering could be divided into two categories: N and U [22]. An N process happens when two of the phonon wave vectors are small the third wave vector is just their summation falling inside the Brillouin zone (BZ), it conserves the total crystal momentum and induces no thermal resistance by itself, and merely redistributes momentum among different phonon modes. While in a U process two of the phonon wave vectors are large, their summation will fall outside the BZ and reciprocal lattice vectors have to be involved to bring it back. These cause change in the total momentum and leads to thermal resistance. N processes do not lead to thermal resistance themselves but contribute indirectly to K by redistributing overall phonon population which can further participate in U process. Therefore, neglecting N processes provides a false prediction of K and must be captured carefully since simply adding the N and U process together ignores the special role of N process.

Figure 2.1: Scattering mechanism shown on a temperature v/s conductivity plot. The thermal conductivity is mainly contributed by boundary scattering under low temperature, while phonon-phonon scattering, and impurity scattering dominate at high temperature.
More specifically, conservation of momentum for three phonon interactions can be written as \( q \pm q' = q'' + G \), where \( G \) is reciprocal lattice vector. For an N process \( G=0 \) and \( G \neq 0 \) for U process. Similarly, for the conservation of energy the absorption process results in one phonon with the combined energy of two incident phonon \( (\omega_\lambda + \omega'_\lambda = \omega''_\lambda) \), while in the decay process the energy of one incident phonon is split into two phonons \( (\omega_\lambda = \omega'_\lambda + \omega''_\lambda) \). In the next section we discuss more about the three-phonon scattering processes which is an intrinsic scattering property of the material.

2.2 Phonon-Phonon Scattering

Broido et al. [29] developed an ab initio approach combining first-principles calculations for harmonic and anharmonic IFCs using DFPT and solving the BTE iteratively, achieving excellent agreement with experiments. Applying this parameter free approach on Si and Ge, excellent agreement with experimentally measured K valid over a temperature range
around room temperature is achieved. However, this approach is computationally expensive.

For a small temperature gradient $\nabla T$, the phonon BTE can be linearized with

$$n_\lambda = n^0_\lambda + n^0_\lambda (1 + n^0_\lambda) \varphi_\lambda$$

where $\varphi_\lambda = \frac{n_\lambda}{n^0_\lambda (n^0_\lambda + 1)}$. The linearized BTE for a perfect bulk crystal free of defects or impurities can be written as

$$v_\lambda \frac{\partial n^0_\lambda}{\partial T} \nabla T = \sum_{\lambda'\lambda''} \left[ \Gamma^+_{\lambda\lambda'\lambda''}(\varphi_\lambda' - \varphi_\lambda'' - \varphi_\lambda) + \frac{1}{2} \Gamma^-_{\lambda\lambda'\lambda''}(\varphi_\lambda' + \varphi_\lambda'' - \varphi_\lambda) \right] \quad \ldots (10)$$

where the left-hand side of equation represent phonon diffusion induced by the thermal gradient and the right hand side represent the total scattering rate of phonon due to three phonon interactions. In the above equation $v_\lambda$ represents the phonon velocity, $\Gamma^\pm_{\lambda\lambda'\lambda''}$ are the three phonon scattering rates which are calculated using first principles. The sums on the right-hand side of equation are over the phase space of energy and momentum conserving normal and Umklapp three-phonon process. The presence of $\varphi_\lambda'$ and $\varphi_\lambda''$ conveys the elastic nature of phonon – phonon scattering. The scattering rate of phonons due to three-phonon process can be determined from Fermi’s golden rule, where the transition rates under the linearized BTE formalism [17,18,19] is expressed as:

$$\Gamma^+_{\lambda\lambda'\lambda''} = \frac{h}{4} \frac{n^0_\lambda - n^0_\lambda'}{\omega_\lambda \omega_{\lambda'} \omega_{\lambda''}} |V^+_{\lambda\lambda'\lambda''}|^2 \delta(\omega_\lambda + \omega_{\lambda'} - \omega_{\lambda''}) \quad \ldots (11)$$

$$\Gamma^-_{\lambda\lambda'\lambda''} = \frac{h}{4} \frac{n^0_\lambda + n^0_\lambda'}{\omega_\lambda \omega_{\lambda'} \omega_{\lambda''}} |V^-_{\lambda\lambda'\lambda''}|^2 \delta(\omega_\lambda - \omega_{\lambda'} - \omega_{\lambda''}) \quad \ldots (12)$$

To successfully compute the values of $\Gamma^\pm_{\lambda\lambda'\lambda''}$ we need the three-phonon interaction matrix element $|V^\pm_{\lambda\lambda'\lambda''}|^2$ measuring the strength of scattering events given by

$$V^\pm_{\lambda\lambda'\lambda''} = \sum_p \sum_{qr} \sum_{\alpha\beta\gamma} \varphi_{pqr} e_{qj}(p) e_{qj}(p) e_{p\pm q}(r) e_{p\pm q}(r) \sqrt{M_p M_q M_r} \quad \ldots (13)$$
In the sums, p only runs over the atoms in what we shall call the central unit cell, but q and r cover the whole system \( r_p^\alpha \) and \( M_p \) denote the \( \alpha \) component of the displacement from the equilibrium position and the mass of the pth atom, respectively. The

\[
\phi_{pqr}^{\alpha \beta \gamma} = \frac{\partial^3 E}{\partial r_p^\alpha \partial r_q^\beta \partial r_r^\gamma}
\]

which is the 3\textsuperscript{rd} order anharmonic IFC. In these expressions, p, q and r run over atomic indices while \( \alpha, \beta \) and \( \gamma \) denote Cartesian coordinates. The third order an-harmonic IFCs, \( \phi_{pqr}^{\alpha \beta \gamma} \) are determined using the 2n+1 theorem [15]. This theorem states that if the derivatives of the wave function up to order \( n \) are known, then it is possible to calculate the energy derivatives for the system up to order \( 2n+1 \).

Equation (10) is solved using the iterative approach [6] to obtain the nonequilibrium distribution functions, \( \phi \lambda \). These are then used to calculate the phonon thermal conductivity tensor \( K_{ij} \). It allows us to obtain the numerical solution for the real crystal at any desired degree of accuracy, the problem being only reduced to the computing time required to reach, correspondingly, a satisfactory order of iteration.

2.3 Models for calculating thermal conductivity

Proper treatment of N scattering and U scattering holds the key to accurately model phonon transport. The iterative numerical approach discussed above gives no relative weight to N and U processes and is completely independent of any assumptions, but it takes a long time to reach the satisfactory order of iteration and is limited to simple materials [6].

2.3.1 Relaxation time approximation

A more affordable approach appropriate is to model phonon-phonon interactions using the relaxation time approximation (RTA), where the relaxation time represents how fast
the non-equilibrium distribution functions return to the equilibrium due to scatterings, hence are called the relaxation times [7]. The scatter term, using RTA can be expressed as

\[ \frac{\partial n_{\lambda}}{\partial t} = - \frac{n_{\lambda} - n_{\lambda}^0}{\tau_{\lambda}^{RTA}} \]  

(14)

where the relaxation time includes phonon-phonon scatterings. These scattering terms linearly depend on the deviations of distribution functions from equilibrium and are solved combined with BTE. The phonon conductivity tensor within RTA is expressed as

\[ K_{ij}^{RTA} = K_D = \frac{\hbar^2}{N\Omega k_B T^2} \sum_{\lambda} \omega_{\lambda}^2 v_{\lambda i} v_{\lambda j} \tau_{\lambda}^{RTA} n_{\lambda}^0 [n_{\lambda}^0 + 1] \]  

(15)

where the \( N\Omega \) represent crystal volume.

The estimation of phonon-phonon scattering in theoretical calculations is a challenging task as it requires fitting parameters and approximations [27], and therefore for our analysis we are using the experimentally measured \( K \) for the highly enriched crystal containing 99.995% of Si\(^{28}\) as a benchmark test data. The measured \( K \) in work [28] provides us with the most accurate approximation of the intrinsic thermal conductivity of single crystal silicon, determined solely by the diffusive boundary and anharmonic phonon processes scattering over a wide temperature range. In theoretical calculation, estimation of phonon-phonon scattering is especially a challenging task as it requires many fitting parameters and approximations, hence experimental data on \( K \) of a single-isotope crystal is valuable as they represent almost solely the phonon-phonon process over a wide temperature range. The \( K \) calculated using iterative and RTA for Si between 10 K and 425 K is shown in Figure 2.1.

Above 80 K, comparing the \( K \) we find good agreement with the experimentally measured results. However, at low temperature we see that iterative and RTA results give
poor approximations for thermal conductivity. To understand this we note that the role of N process consists of transferring heat between groups of modes while the U process dissipate it, therefore when there is no U scattering K must diverge because N scattering cannot provide thermal resistance as seen in iterative calculations. This divergence however does not occur in RTA because it treats both N and U with same weight and both enter in the scattering rate in a purely resistive manner [8,9,10]. This is equivalent to stating that any time a single phonon is scattered a given amount of heat current is dissipated, which contradicts the momentum-conserving character of the N process. Since, not all the scattering processes are completely resistive, RTA underestimates K [27]. The method is good for describing thermal transport in 3D crystal structures at temperatures above room temperature, where the high number of scattering events assures an isothermal repopulation

Figure 2.3: Thermal conductivity of isotopically pure Si calculated with iterative and RTA method, as a function of temperature. Blue star shows experimentally measured results [28]. The black and green line indicates results from iterative method and RTA method using ShengBTE.
of the phonon modes, while its use becomes questionable in the range of low temperatures, or in systems where the U process are frozen out and N processes dominate the phonon relaxation [5,11,12]. To correct for this, a new model was proposed by Callaway to separate the effects of N scattering and U scattering on heat transport [5].

2.3.2 Callaway Model

The Callaway model has been extensively used in phonon transport studies because it preserves the efficiency of the RTA while significantly approaching the accuracy of the exact solution schemes [24]. As discussed in previous sections, RTA gives a finite $K$ in case of N processes alone, hence it underestimates $K$. Callaway proposed an intuitive model to treat N and U separately [5] which can be written as

$$\frac{\partial n_{qj}}{\partial t} = -\frac{n_{qj} - n^0_{qj}}{\tau_U} - \frac{n_{qj} - n_{qj}^{\text{disp}}}{\tau_N} \ldots (16).$$

Where the first term on the right-hand side of the equation represents the rate at which the nonequilibrium distribution returns to the equilibrium distribution due to momentum destroying resistive scattering mechanism. The second term represents how the perturbed distribution function in the presence of collective momentum conserving normal phonon-phonon processes relaxes to a drifted distribution $n_{\lambda}^{\text{disp}}$, often referred to as the flowing equilibrium. The flowing equilibrium is written as

$$n_{\lambda}^{\text{disp}} = \frac{1}{\exp\left[\frac{\hbar \omega_{\lambda} - Q \cdot \Lambda}{k_B T}\right] - 1}$$

where $\Lambda$ is a Lagrange multiplier enforcing momentum conservation and the terms $\tau_U$ and $\tau_N$ represent the U and N scattering rates. To solve $\Lambda$, Callaway utilized the fact
that N process conserve phonon momentum. The rate of change of the total phonon momentum due to N process is set equal to zero.

\[
\sum_{\lambda} \frac{n_{\lambda} - n_{\lambda}^{\text{disp}}}{\tau_N} q = 0
\]

The Callaway thermal conductivity can be written as

\[
K_{ij}^{\text{Callaway}} = K_D + \frac{\lambda_1 \lambda_2}{\lambda_3} \ldots (17)
\]

The correction terms are expressed as

\[
\lambda_1 = \lambda_2 = \frac{1}{N\Omega k_B T^2} \sum_{\lambda} \hbar^2 \omega_\lambda v_\lambda q_\lambda \frac{\tau_{\text{RTA}}}{\tau_N} n_{\lambda}^0 [1 + n_{\lambda}^0]
\]

\[
\lambda_3 = \frac{1}{N\Omega k_B T^2} \sum_{\lambda} \frac{q_\lambda q_\lambda}{\hbar^2 \omega_\lambda} \frac{\tau_{\text{RTA}}}{\tau_N \tau_U} n_{\lambda}^0 [1 + n_{\lambda}^0]
\]

Here, using the empirical form for \(\tau_N\) and \(\tau_U\) from equation (7) and (8) we determine the scattering rates analytically and calculate the K of Si using Callaway model. While the Callaway approach uses empirical rates, it lacks predictive ability and is not as exact as the iterative and direct BTE solver method, it is widely used to fit the observed thermal conductivities in terms of boundary scattering, point-defect scattering and U processes in a number of cases, notably diamond [20], lithium fluoride [21,22] and has shown accurate results in graphene [5].

Our results in Figure 2.2 for the calculated value of K of Si using Callaway model shows that using empirically calculated rates at low temperature the model overestimates thermal conductivity while at high temperature around room temperature it underestimates K. We see a large deviation in calculated K value over the entire temperature range which suggests that we need to calculate the rates used in the calculations more accurately.
2.3.3 Improved Callaway Model: Allen Model

The major advantage of Callaway model is its elegant method allowing us to handle the summation of both momentum-conserving N-processes with non-conserving U-processes, whose relaxation times are not additive in a straightforward manner. An improvement in Callaway model was proposed by Allen which indicated that Callaway model underestimated the suppression of N process. A different constraint to solve for \( \Lambda \) is used, according to which the total phonon momentum should be same for both actual and displaced equilibrium distribution. \( \hbar^2 \omega_\lambda v_\lambda q_\lambda \)

\[
\sum_\lambda (n_\lambda - n_{\lambda}^{\text{disp}}) q = 0
\]

The improved Callaway model modified the correction terms used in the Callaway \( \tau^\text{RTA}_\lambda \) which are expressed as

\[
\lambda_1 = \frac{1}{N\Omega k_B T^2} \sum_\lambda \hbar^2 \omega_\lambda v_\lambda q_\lambda \tau^\text{RTA}_\lambda n_\lambda^0 [1 + n_\lambda^0]
\]

\[
\lambda_2 = \frac{1}{N\Omega k_B T^2} \sum_\lambda \hbar^2 \omega_\lambda v_\lambda q_\lambda \tau^\text{RTA}_\lambda \tau_N n_\lambda^0 [1 + n_\lambda^0]
\]

\[
\lambda_3 = \frac{1}{N\Omega k_B T^2} \sum_\lambda \frac{q_\lambda q_\lambda}{\hbar^2 \omega_\lambda} \tau^\text{RTA}_\lambda \tau_U n_\lambda^0 [1 + n_\lambda^0]
\]
The improved Callaway model showed that using the modified correction terms and simple parametrization for $\tau_N$ and $\tau_U$ can lead to higher K. Similar calculations of K shown in Figure 2.4 was performed using the improved Callaway model, where we observe a better agreement in calculated K of Si above 125 K as compared to the Callaway model. However, for temperature below 80 K we see the improved Callaway model overestimates K.

### 2.4 Conclusion

From our results we see that at low temperature the Callaway and the improved Callaway model show good agreement with results where boundary scattering is dominant, whereas
at high temperature iterative and RTA models show good agreement with the experimentally measured thermal conductivity where phonon-phonon scattering is dominant. Hence, there lies a need for a model which can characterize K properly at low and high temperature. We see a good agreement in the calculated room temperature K from RTA and iterative is 155 [W/m-K] agreeing with the experimental results. However, using Callaway model and improved Callaway model a lower value of thermal conductivity 103 [W/m-K] and 124 [W/m-K] with underestimation of 34% and 20% is observed. This demonstrates that neither the original Callaway model nor improved Callaway model has guaranteed accuracy, in the case of Si. It can either overestimate or underestimate K, hence, a systematic examination is still lacking to accurately capture the three-phonon scattering over a wide temperature range.
CHAPTER 3

THERMAL TRANSPORT IN SILICON

From our previous results we see that at very low temperatures the calculated and experimentally measured lattice thermal conductivities are in good agreement. For temperatures above the peak, the calculated thermal conductivity becomes significantly larger than the measured value. The discrepancy in results suggests that a proper treatment of the phonon-phonon scattering rate is missing.

3.1 Previous Modelling Work

Phonon-phonon interactions is the main contribution to the thermal resistance for semiconductors in most temperature ranges. Using RTA we write scattering term as

\[
\frac{\partial n_\lambda}{\partial t} = - \frac{n_\lambda - n_\lambda^0}{\tau_{\lambda}^{RTA}}
\]

where the relaxation times are defined for all the different phonon scattering mechanisms. The RTA treats all mechanism as resistive process however N process are non-resistive process. The observation that N and U process need to be treated separately was addressed by Callaway. The Callaway model adds a correction term to the existing RTA model given by equation (17). Figure 3.1 show a comparison of the RTA and Callaway model where we see that the RTA part evaluated from Callaway model using analytically computed rates lacks accuracy as the three phonon rates used are fitted from existing experimental data and lacks predictive power and transferability over a range of temperature. This suggests that we need to accurately capture the three-phonon scattering rates.
Figure 3.1: Thermal conductivity comparison for Si RTA vs Callaway. Green line shows RTA calculations from first-principles calculations, dotted blue shows RTA part from empirical Callaway model, and dotted red line shows correction part of empirical Callaway model. We see here a 31% error in the RTA vs empirical Callaway RTA part at 300K.

3.2 Methodology

3.2.1 Phonon Dispersion

Phonon transport properties including frequencies and velocities are largely determined by 2\textsuperscript{nd} order IFC. Therefore, a precise representation of the full phonon dispersions and modes and of the anharmonic forces is required to calculate accurately the K. Around room temperature majority of heat is carried by the dispersive acoustic phonons. The acoustic phonon frequencies and group velocity are important characteristics that will be needed in calculating the K, which is obtained from dispersion and must be described precisely over the entire FBZ. Furthermore, the large wavevector acoustic phonons are involved in the three-phonon U scattering processes that limit the thermal conductivity. Hence, the
acoustic phonon dispersions near the Brillouin-zone boundary must be accurately determined. Calculation of phonon dispersion of Si using first principles Density Functional Perturbation Theory (DFPT) was done using open-source software Quantum-ESPRESSO [33]. Figure 3.2 shows the dispersion and density of states (DOS) relation for Si plotted along the high symmetry path. Using the dispersion data, we further calculate the specific heat which is shown in the Figure 3.3. The slope of the dispersion curves represents the group velocity of phonons. The phonon group velocities are then calculated from the dispersion data using the central difference method. Branch-wise phonon velocities is shown in Figure 3.4. The higher lying optical phonons have low velocities and are high in energy, thus they act as energy storage and do not contribute greatly to heat flow. Conversely, acoustic phonons are high in velocity and are present at much lower temperatures, thus acoustic phonons are the main contributors to thermal transport.

Figure 3.2: Phonon Dispersion of Si along symmetry path and DOS
Figure 3.3: Specific Heat of Silicon

Figure 3.4: Branch wise phonon velocity of Silicon
3.2.2 Three Phonon Scattering

From the works of Callaway, Kelemen, Holland [32] the relaxation time due to three-phonon scattering can be expressed as

\[
\frac{1}{\lambda} = A \omega^n f(T) \quad \ldots (19)
\]

where \( n \) is an integer, \( \omega \) phonon frequency and \( f(T) \) is one of the several temperature dependent functions and \( A \) is constant determined by fitting to the experimental thermal conductivity data. For the derived relaxation time expression (18) modelling was done assuming small phonon frequencies and low temperature also the three-phonon scattering strength was modeled taking all three acoustic phonons to have low frequency, an assumption that is incompatible with the U scattering processes that produce the thermal resistance. Since this approach is not consistent to describe the high temperature thermal transport it needs to be assessed more. Ward et al. [32] derived correct forms of intrinsic phonon relaxation times describing N and U phonon-phonon scattering around and above room temperature from first principles. The relaxation time frequency dependence with respect to the total relaxation time is shown for Si in Figure 3.5, which highlights the importance of using the correct scaling behavior of the relaxation time for the three-phonon scattering mechanism, which directly affects the estimation of the thermal conductivity.

From our results we see that all the models show good agreement with the experimentally measured thermal conductivity at high temperature. We see discrepancies in result around room temperature where the dominant scattering is from N process. N scattering usually dominates for relatively low-frequency phonons, while the U process starts to dominate at higher frequencies, giving rise to a higher-order frequency dependence. For a deeper analysis of heat transfer correct scaling behavior of N and U
process is important as three-phonon scattering mechanism directly affects the estimation of the thermal conductivity [32]. Callaway and improved Callaway models use approximations to represent the phonon properties, as these cannot be easily measured experimentally or simulated numerically, hence lacks guaranteed accuracy for calculation of thermal conductivity. Using ab-initio calculated three-phonon scattering rates Jinlong Ma et al [27] studied the K of Si with the Callaway and improved Callaway model and showed satisfactory agreement with the exact ab-initio results for room temperature. However, for temperature at 50 K the RTA gives about 24% underestimation and both the original Callaway model and Allen modified model underestimate thermal by 19% and
11% respectively. This suggests that an accurate calculation of N and U scattering rates is needed for accuracy of K calculations.

### 3.2.2.1 Calculation of Normal and Umklapp Scattering Rates

In order to fully capture the N and U scattering rates and their frequency dependence accurately, we calculate them using the first principles over a wide temperature range 10 K to 425 K for perfect bulk Si with zero isotopic scattering. The second and the third order of IFCs used in the calculations was obtained from almaBTE [36]. All simulations are performed using software package ShengBTE [2]. A 17x17x17 qpoint grid was used and the cubic lattice parameter used is 5.47Å. The ShengBTE is a software package is implemented in Fortran. The source code has multiple subroutines which are parallelized using MPI. These parallelized subroutines are executed in an interleaved manner thus making the task of modifying and debugging the source code a highly challenging and involving task. The conditions of the N and U scattering process discussed in the previous section 2.1 were implemented as a subroutine in the ShengBTE software package by me and separated from the total scattering rate as

\[
\frac{1}{\tau} = \frac{1}{\tau_U} + \frac{1}{\tau_N}
\]

Though there exists DFT packages which can compute the total scattering but to the best of our knowledge there is no package which implements the computation of converged sets of ab-intio N and U phonon scattering rate. The rates are then obtained for a set of 165 irreducible q points which are then expanded across set of 4913 q points covering the entire FBZ and following which interpolated on a dense grid of 32000 k-points.
In Figure 3.6 we show the N scattering rates for different temperature and how their frequency dependence varies as the temperature increases. At low temperature of 50 K and 100 K we see $\omega^3$ and $\omega$ dependence, while for higher temperature 200 K and 300 K we see $\omega^2$ dependence. Similar, observation was done for U scattering rates shown in Figure 3.7 where we see $\omega^3$ dependence at 50K while $\omega^4$ dependence at 100 K, 200 K and 300 K, respectively. Hence, we conclude that a single power cannot be a great fit for entire temperature range.

Figure 3.6: Normal scattering from first principles at various temperature with respect to phonon frequency. The scattering rates are fitted to frequency where the yellow dots represent scattering rate for 50 K seeing a $\omega^3$ dependence. The red and blue dots represent scattering rate for 200 K and 300 K respectively where we see $\omega^2$ dependence.
Figure 3.7: Umklapp scattering from first principles at various temperature with respect to phonon frequency. The scattering rates are fitted to frequency where the yellow dots represent scattering rate for 50 K seeing a $\omega^3$ dependence. The magenta, red and blue dots represent scattering rate for 100K, 200 K and 300K respectively where we see $\omega^4$ dependence.

3.3 Thermal Conductivity Calculations

We calculate the $K$ using equation (3) of bulk Si sample of cross section 4.35x4.35 mm$^2$ and a length of 40 mm with average surface roughness of 0.2 μm across a wide temperature range of 10 K to 425 K. The inputs required are the phonon dispersions, velocities and N and U scattering rates which are obtained using the methodology discussed in the previous sections. Using the scattering rates for N and U obtained from first principles in the Callaway and improved Callaway model, we see satisfactory agreement in the $K$ with the experimentally measured $K$ value which is shown in Figure 3.8. The calculated $K$ of Callaway and improved Callaway model at room temperature are 151.2 [W/m-K] and 156.9 [W/m-K] which agree with the experimentally measured value 155 [W/m-K]. This
demonstrates that using first principles calculated N and U scattering rates we captured the proper scaling and frequency dependence of the three-phonon scattering process for K calculations. Also, we can say that in case of Si the improved Callaway model and Callaway model both performs equally well.

![Thermal conductivity of Si](image)

**Figure 3.8:** Thermal conductivity of Si calculated from Callaway and improved Callaway model (Allen model) where the scattering rates are computed analytically and using first principles. Blue star shows experimentally measured results [28].

### 3.4 Thermal Conductivity Calculations in Silicon Nanowire

Our results in the previous section demonstrates that using first-principles calculated rates we can accurately capture the K for a bulk Si. Therefore, using the same technique, we extend our results to study the K of Si nanowire (NW). We perform K calculation along transport direction <100>, for a diameter of 20 nm, 35 nm and 55 nm. Figure 3.9 shows a plot of K of Si NW of different diameters. From our calculations we observe that the calculated K of a short length Si NW is two order of magnitude lower than that of bulk Si,
and as we reduce the wire diameter, we see a decrease in K as well. This indicates that boundary scattering is dominant in Si NW.

Figure 3.9: Thermal conductivity of Si nanowire for different diameter sizes calculated from Callaway model using first principles scattering rates.

Further, for short length Si NW we observe peaks around 200 K, while in case of bulk Si we observe a clear peak at 25 K. The shift of peak suggests that, as the wire diameter is reduced the boundary scattering dominates over three-phonon scattering. At low temperature there is mostly boundary scattering so the K rises in proportion to the diameter, while at high temperature phonon-phonon scattering also exists so K rises more slowly than diameter. Figure 3.10 shows us that as we increase the diameter of a Si NW from 100 nm to 500 nm, K increases by a factor of 4 at low temperature where boundary scattering is dominant and less than 4 at high temperature where three-phonon scattering is dominant,
therefore the peak gradually shows as we increase the diameter indicating that boundary scattering is dominant in case of NW.

![Graph showing thermal conductivity for different diameters](image)

**Figure 3.10:** Thermal conductivity of Si nanowire for length of 1mm and different diameter calculated from Callaway model using first principles scattering rates.

### 3.5 Conclusion

The analytical expressions given by Callaway model have served as a valuable tool in the field of semiconductor heat transfer for many years. We have demonstrated that using the analytically calculated scattering rates provides a poor representation of the actual solution of BTE because of the improper scaling behavior for N and U process. In this work, we calculate the thermal conductivity of silicon using a hybrid model approach where we obtain the N and U scattering rates from first principles and implement it in the Callaway model. Using this approach, we see good predictive abilities as it uses no adjustable parameters and is computationally inexpensive. We showed here that the
Callaway model can be used to describe the thermal conductivities of silicon for temperatures ranging from 10 K to 425 K with good agreement, using the anharmonic scattering rates for N and U process calculated from first principles.
CHAPTER 4
THERMAL TRANSPORT IN DIAMOND

Diamond has the highest K than any known bulk material, its ability to conduct heat far surpass that of commonly used semiconductor material Si. Unlike convection-based cooling fans and heat sinks, diamond films can be directly integrated into a chip which cools much more efficiently. With the room temperature K value of 3000 [W/m-K], integration of diamond into GaN HEMT ultra high-power devices is a good option to optimize thermal management, enhancing the device reliability and thus expanding their lifetime.

4.1 Overview of Phonon Transport in Diamond

The dominance of N scattering in diamond is noticed previously [36]. In materials such as Si, U scattering is strong and the converged solution of phonon BTE is less than 10% higher than RTA solution as the behavior of K is dominated by U scattering. However, in diamond the U scattering is weaker than Si and the contribution of N process is larger, which manifests itself in the converged solution of the phonon BTE giving a K that lies far higher than RTA. In RTA we do not see any divergence because the N and U process are independent and both enter in a purely resistive manner in the total scattering rate, however in the iterative procedure if there is no U scattering and N process are included, the lack of thermal resistance manifests which suggests there is a large normal correction term for diamond and correction to the normal contribution is more important here.

For diamond, the strong bond stiffness and light atomic mass produce extremely high phonon frequencies and velocities, which results in extremely large K. Diamond when
compared to other common semiconductors also has a small phase space for U scattering [34]. These quantities enter directly in the thermal conductivity calculations and so contribute to its increasingly high K compared to other materials. The plot of phonon frequency and velocities is shown in Figure 4.1 and Figure 4.2, respectively. Further, we calculate the specific heat from the dispersion data shown in Figure 4.3, from the figure we observe that heat capacity does not level off until above room temperature. Diamond has a high $\theta_D$ $2240$ K and around $\theta_D$ the heat capacity flattens out as most modes are thermally excited. Also, above $1/3$ of $\theta_D$ U scattering takes over because we need large q phonons for U scattering, and they are not excited very much below $\theta_D$ and at those temperature N scattering plays a larger role. Therefore, for materials having high $\theta_D$ typically have a lots of N scattering.

Figure 4.1: Phonon Dispersion of diamond along symmetry path and DOS
Figure 4.2: Branch wise phonon velocity of Diamond

Figure 4.3: Specific Heat of Diamond
4.2 Thermal Conductivity Calculations in Diamond

We calculate the $K$ of a bulk diamond for a temperature range of 100 K to 425 K using first-principles. The Figure 4.4 shows a plot of $K$ as a function of temperature for different models. From the plot we observe that iterative solution gives a higher value of $K$ as compared to RTA, for instance 9% larger at room temperature. This underestimation in RTA is because of the equally resistive treatment of N process, which become stronger at low temperature. Also, for Callaway and improved Callaway we see an error of 48% and 40% respectively. Figure 4.5 shows the N correction observed from the respective models and suggests that further development is required in the Callaway and improved Callaway model to capture the N correction properly.

![Figure 4.4: Thermal conductivity calculations of diamond using iterative, RTA, Callaway and improved Callaway model.](image)

Figure 4.4: Thermal conductivity calculations of diamond using iterative, RTA, Callaway and improved Callaway model.
Figure 4.5: Normal correction term from different models calculated thermal conductivity for diamond

4.3 Conclusion

In case of Si where the N correction was small, we were successfully able to capture the K accurately over a wide temperature range using the Callaway and the improved Callaway model. However, in case of diamond, where N scattering is dominant the correction from N process is not accurately captured by the Callaway and improved Callaway model and needs further investigation. In order to evaluate the K accurately for any material we need capture the N and U process better by using the first-principles calculated N and U scattering rates into our hybrid model rather than using analytical rates calculated using approximations.
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