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Multiscale Nondiffusive Heat Transfer in Dielectric Materials

by

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To the unexpected journey I had here.
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Curriculum Vitae

Education


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Nomenclature and Acronyms

Roman Letters

\(a\) lattice constant, m
\(\Delta h\) surface deformation, m
\(\Delta t\) temporal step, s
\(\Delta x, \Delta y, \Delta z\) spatial step, m
\(\hat{s}\) unit directional vector (vector), 1
\(\hat{x}, \hat{y}, \hat{z}\) unit directional vector (vector) for \(x, y, z\) respectively, 1
\(\mathbf{q}_{aw}\) wall heat flux (vector), W/m\(^2\)
\(\mathbf{v}\) velocity (vector), m/s
\(c\) speed of sound, m/s
\(c_s\) Debye velocity, m/s
\(c_v\) volumetric specific heat, J/m\(^3\)K
\(c_v\) volumetric specific heat, J/m\(^3\)K
\(D\) phonon density of states, 1
\(E\) volumetric energy density, J/s\(^3\)
\(e\) Volumetric energy density per solid angle, J/m\(^3\)
\(e^0\) equilibrium phonon energy density, J/m\(^3\)
\(e_{\omega}\) volumetric energy density per unit frequency and solid angle, J/s/m\(^3\)
\(f\) density distribution function, 1
\(f^0\) equilibrium distribution function, 1
\(H\) optical signal, 1
\( h_c \) convective heat transfer coefficient, \( \text{W/m}^2\text{K} \)

\( I \) light intensity, \( \text{W/m}^2 \)

\( k_B \) Boltzmann constant, \( \text{m}^2\text{kg/s}^2\text{K} \)

\( Kn \) Knudsen number, 1

\( L \) width of metal heater, m

\( L_c \) system characteristic length, m

\( N_A \) Avogadro’s number, 1

\( P \) grating period size, \( \mu\text{m} \)

\( r \) interface resistivity, \( \text{Km}^2/\text{W} \)

\( s \) Laplace transform variable in time, \( \text{s}^{-1} \)

\( T_w \) wall temperature, K

\( T_{\infty} \) environmental temperature, K

\( v_p \) group velocity, m/s

\( x, y, z \) spatial coordinates, m

\( \text{Kn} \) Knudsen number, 1

\( E \) volumetric energy density, \( \text{J/m}^3 \)

\( \text{F} \) force (vector), N

\( f_{BE} \) Bose-Einstein statistics, 1

\( \text{G} \) reciprocal lattice vector (vector), \( \text{m}^{-1} \)

\( h \) Planck constant, \( \text{J/s} \)

\( \hbar \) reduced Planck constant, \( \text{J/s} \)

\( k \) wavevector (vector), \( \text{m}^{-1} \)

\( K \) spring constant, \( \text{N/m} \)

\( m \) mass, kg

\( p \) momentum (vector), \( \text{N/s} \)

\( q \) heat flux (vector), \( \text{W/m}^2\text{K} \)
\(Q\)  volumetric heat generation rate, W/mK

\(r\)  position (vector), m

\(T\)  temperature, K

\(t\)  time, s

\(u_j\)  displacement of \(j^{th}\) atom, m

\(V\)  volume, m³

**Greek Letters**

\(\beta\)  nonequilibrium absolute temperature, K

\(\alpha_h\)  coefficient of linear thermal expansion of heater, K\(^{-1}\)

\(\alpha_s\)  coefficient of linear thermal expansion of substrate, K\(^{-1}\)

\(\beta_1\)  absorption coefficient in pump laser, m\(^{-1}\)

\(\beta_2\)  absorption coefficient in probe laser, m\(^{-1}\)

\(\delta_{AD}\)  average difference, 1

\(\gamma\)  thermal decay rate, s\(^{-1}\)

\(\lambda\)  wavenumber, m\(^{-1}\)

\(\lambda_e\)  laser wavelength, m

\(\mu\)  directional cosine, 1

\(\nu_s\)  Poisson ratio, 1

\(\Omega\)  solid angle, 1

\(\Omega_D\)  Debye cutoff frequency, s\(^{-1}\)

\(\omega_i\)  weighting factor in lattice Boltzmann method, 1

\(\omega_r\)  weight coefficient for successive overrelaxation, 1

\(\rho\)  mass density, kg/m³

\(\tau_c\)  characteristic time for ballistic phonon transport, s

\(\Theta_D\)  Debye temperature, K
\( \xi \) Fourier transform variable in space, m\(^{-1}\)

\( \zeta \) nondimensional number that characterizes the nondiffusive effects, l

\( \kappa \) thermal conductivity, W/mK

\( \Lambda \) phonon mean free path, m

\( \Lambda_B \) ballistic phonon mean free path, m

\( \Lambda_D \) diffusive phonon mean free path, m

\( \omega \) phonon angular frequency, s\(^{-1}\)

\( \phi \) azimuthal angle, l

\( \phi \) azimuthal angle, l

\( \tau \) relaxation time, s

\( \theta \) polar angle, l

\( \tau_B \) ballistic phonon lifetime s

\( \tau_D \) diffusive phonon lifetime, s

**Abbreviations**

1D one-dimensional

2D two-dimensional

3D three-dimensional

AMM acoustic mismatch model

BDE Ballistic-diffusive equation

BGK approximation Bhatnagar-Gross-Krook approximation

BTE Boltzmann transport equation

C-V model Cattaneo-Vernotte model

DFT density function theory

DMM diffusive mismatch model

EG-BTE Enhanced Gray Boltzmann transport equation

EPRT equation of phonon radiative transfer
ETC  effective thermal conductivity
FD  finite difference
G-K model  Guyer-Krumhansl model
ILBM  implicit lattice Boltzmann method
LA  longitudinal acoustic
LBM  lattice Boltzmann method
LHS  left hand side
LO  longitudinal optical
MD  Molecular Dynamics
MFP  mean free path
MG  metallic grating
N process  normal process
NILT  numerical inverse Laplace transform
PDE  partial differential equation
RHS  right hand side
RTE  radiative transfer equation
TA  transverse acoustic
TDTR  time-domain thermoreflectance
TO  transverse optical
TPHC model  Two-Parameter Heat Conduction model
TTG  transient thermal grating
U process  Umklapp process
Abstract

Multiscale Nondiffusive Heat Transfer in Dielectric Materials

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With the progress of nanotechnologies for microelectronic and thermoelectric devices, a lack of understanding of the heat transfer in nano-/micro-scale creates the bottleneck for the applications. Although Fourier’s law is able to describe the heat transfer in macroscale, it breaks down in nano-/micro-scale due to the nondiffusive heat transfer, which is still not well understood. Consequently, the objective for this thesis is to achieve a fundamental understanding of nondiffusive heat transfer.

Phonon is a quantum mechanical description of lattice oscillations, and it is the main heat carrier in insulators and semiconductors. When the system sizes are comparable or even smaller than the mean free path (MFP) of phonons, the heat transfer becomes nondiffusive due to the contribution from ballistic phonon transport. The phonon-mediated heat transfer in nanosystems and their interfaces is very complicated. In this thesis, we mainly focus on three problems: (1) there lacks a criterion to evaluate when Fourier’s law breaks down; (2) the size-dependence of thermal interface resistivity is not clearly captured; (3) the ballistic heat transfer has not been directly observed at room temperature.

Accordingly, there are three goals for this thesis: (1) to find the criterion which is able to identify when Fourier’s law breaks down; (2) to characterize how interface resistivity changes with the hotspot size; (3) to seek the possibility of a direct observation of ballistic heat transfer at room temperature.

To achieve the first goal, a criterion is proposed based on the nondimensional parameter defined in the Two-Parameter Heat Conduction (TPHC) model, to predict the breakdown of Fourier’s law. The physical interpretation of this nondimensional parameter is a product of the nondimensional diffusive phonon MFP and ballistic phonon MFP. To validate this criterion, both Fourier’s law and the TPHC model are analytically or numerically solved for multiple experiments on different materials. These experiments include the one-dimensional and two-dimensional transient thermal grating (1D/2D TTG) experiments on silicon and gallium arsenide, and the metallic grating (MG) experiments on sapphire. The comparison results indicate that this criterion is able to identify the breakdown of Fourier’s law.

To achieve the second goal, the TPHC model and the enhanced gray Boltzmann transport equation (EG-BTE) are numerically solved on the MG experiments by finite difference method and lattice Boltzmann method, respectively. Current study of the interface resistivity is based on Fourier’s law, which fails to decouple the size-dependence of the thermal conductivity (material thermal property defined by Fourier’s law) and the interface resistivity. While the TPHC model
and EG-BTE are able to exclude the size-dependence of the thermal conductivity, and thus extract a clean size-dependence of the interface resistivity. Based on these two models, a monotonically change of the interface resistivity with the size of interface is captured, which is different from but more reliable than the results by Fourier’s law.

To achieve the third goal, we focus on the thermal wave, the direct evidence of ballistic heat transfer. Although ballistic heat transfer is believed to be the cause of nondiffusive heat transfer, so far only the deviation from prediction by Fourier’s law is reported as the evidence of nondiffusive heat transfer at room temperature. To study the thermal wave, the EG-BTE is applied on the thermal wave experiments in cryogenic crystal. In these cases, the EG-BTE is numerically solved by an in-house numerical scheme developed called the discrete ordinates method for phonon transport, which is validated by other theoretical study in this thesis. The results are compared with the experiments and indicate a successful reproduction of the thermal wave. Based on the proved capability of describing the thermal wave, the EG-BTE is extensively applied to identify the length and time scale to observe the thermal wave at room temperature.

In conclusion, the criterion proposed in this thesis facilitates the choice of models between the efficient Fourier’s law and the accurate nondiffusive models, benefiting the engineering use in multiscale situations. Secondly, the monotonic behavior of interface resistivity offers new insights of the physics of nondiffusive heat transfer across the interface. At last, the prediction of the thermal wave at room temperature provides potential guidance for the design of experiments to directly observe the thermal wave at room temperature, opening up a new gate to study the unique features of the ballistic heat transfer.
Chapter 1

Introduction

In this chapter, the background for heat conduction is introduced, including both macroscale and microscale descriptions. A great deal of research in heat conduction has been conducted and benefiting the development of engineering technology in recent decades, both experimentally and theoretically. But the physical mechanisms behind heat conduction are still not well-understood. We start from the applications of heat conduction in engineering techniques, such as the micro-electronics cooling, to introduce the background and challenges, followed by a theoretical description of heat conduction. After that, the theory of Boltzmann transport equation is presented, and the state-of-art nondiffusive heat transfer models are reviewed. Then the objectives of this study are listed and explained. In the last section, the outline for this thesis is shown.

1.1 Failure of traditional heat diffusion theory

Heat conduction is one important but common phenomenon. It happens ubiquitously in all phases of matters, including solids, liquids, gases and plasmas. It is the transport of internal energy by the collisions of particles such as molecules, atoms, electrons. It is involved as the key part in numerous engineering applications, including the design of thermoelectric devices and the thermal package design of microelectronics.

Figure 1.1: (a). A schematic of a nanowire thermoelectric device utilizing wasted solar heat. Picture by UCI center for solar energy. (b). An illustrative view of hotspot in microelectronics. Picture by iStock.
Thermoelectric devices show potential to serve as one solution for current energy crisis. At present, roughly 90% of the world’s power is generated by heat engine using fossil fuel with the efficiency to be 30% - 40%. Based on these estimations, roughly $1.5 \times 10^{13}$ W of heat is lost to the environment as waste heat. Furthermore, waste heat is also produced during the consumption processes of generated power. For example, in transportation, the typical energy efficiency is about 20%, which equivalently suggests about $7 \times 10^{11}$ W energy is rejected as waste heat. According to this calculation, a great deal of energy can be produced if a decent ratio of the waste heat can be recycled through some techniques. The application of thermoelectric devices is one of these techniques. The mechanism for a typical thermoelectric device is shown in Fig. 1.1(a). Thermoelectric devices convert the input heat into electricity through Seebeck effect: a temperature gradient in a conducting material results in heat flow; this results in the diffusion of charge carriers. Consequently, this technique closely involves the process of heat conduction.

On the other hand, the thermal package design of microelectronics also requires a deep understanding of heat conduction. With the advancement of the miniaturization technology, transistors, the core part of the microelectronics, are shrinking in size to reach a better portability and higher performance. The transistors are generating heat when working, which is highly localized and accumulated due to the small size (i.e., several nanometers to micrometers) of transistors (Fig. 1.1(b)). The accumulated heat, if not removed efficiently, will finally lead to the thermal failure of the devices. One most straightforward way to remove the accumulated heat is by heat conduction.

Heat conduction traditionally is believed due to the random collisions and diffusion of particles. the kinetic and potential energy, jointly known as internal energy, is gradually dissipating and finally leading to thermodynamic equilibrium. This equilibrium process is characterized by the inhomogeneity of the energy distribution: larger difference in internal energy results in higher energy dissipation rate for two fixed locations in certain material. However, for different materials, the heat conduction rate also varies, even though the same difference exists in temperature, which is defined as a sign of internal energy. Notably, the first empirical relationship between the heat conduction rate in a material and the temperature gradient in the direction of energy flow is formulated by Joseph Fourier, who concluded that “the heat flux resulting from thermal conduction is negatively proportional to the magnitude of the temperature gradient and opposite to it in sign” [1]. Mathematically, it can be expressed as

$$ q = -\kappa \nabla T, \quad (1.1) $$

where $q$ denotes the heat flux vector, $T$ represents temperature, and $\kappa$ is the proportionality constant, defined as the thermal conductivity of the material of interest. In this study, the bold font always denotes a vector quantity. This law, combined with energy conservation, offers solutions to most macroscopic heat conduction problems. Specifically, the thermal conductivity remains roughly a constant for given bulk materials within a certain range of temperature. By this definition, heat conduction is sometimes referred to as “heat diffusion” since it behaves just as mass diffusion.

In macroscopy and at relatively high temperature (i.e., room temperature), the collisions between particles are intense and continuously taking place. One ideal case is to assume the collisions happen everywhere at any moment, which is the diffusive assumption. It is due to the validity of this diffusive assumption that the analysis through Fourier’s law serves macroscopic engineering calculations and predictions with satisfactory precisions. Unfortunately, this assumption does not unconditionally hold. Discrepancies between Fourier’s law predictions and experiments were found
in the miniaturization technology. For example, as shown in Fig. 1.2, the measured effective thermal conductivity (ETC) shows dependence on the system characteristic length $L_c$ instead of remaining as a constant defined by Fourier’s law. Moreover, when $L_c$ approaches the phonon mean free path (MFP), which is defined as the free distance a phonon travels without collision, phonon transport is not diffusive anymore, or even becomes ballistic. The Knudsen number is defined to characterize the heat transport regime: $Kn = \Lambda / L_c$, where $\Lambda$ is the phonon MFP. When $Kn \gg 1$, phonons travel ballistically with few collisions; while $Kn \ll 1$, the diffusive assumption holds since phonons propagate diffusively, experiencing scattering at any moment. Between these two extreme cases, for example $Kn \sim 1$, the size-dependent ETC can be obtained from measurements.

Therefore, it is the decrease in collisions intensity between the heat carriers that leads to the failure of the diffusive assumption, and finally results in the breakdown of Fourier’s law. To better understand the reasons behind it, we need to analyze the heat carriers from a quantum mechanical perspective.

1.2 Main energy carrier in dielectric materials: phonon

In contrast to the simplification by the diffusive assumption, phonons and their behaviors are actually very complicated. Fundamentally, phonons and electrons are two main types of elementary particles or excitations in solids. Both of them transfer heat as energy carriers. Electron is the real particle that owns nonzero rest mass. Its behavior in our length scale of interest is particle-like. It is the main heat carrier in metals and important carriers in some nonmetal materials. In this study, electron is not discussed since we focus on dielectric materials where phonon is the dominant energy carrier. Phonon, on the other hand, is a quantized description of lattice waves. The name of phonon is an analogy to the name of photon, which is a description of electromagnetic radiation waves. Here, we briefly introduce the fundamentals of phonon.
1.2.1 Definition of phonon

When vibrating, the lattice is dissipating energy as traveling waves of various frequencies and corresponding velocities, marked as different modes. A collection of lattice waves can be treated as a group of quasiparticles termed as phonon. According to quantum theory, the eigenvalues or energy levels for the mode $\omega_k$ of a harmonic oscillator are \[ E_n = \left( n + \frac{1}{2} \right) \hbar \omega_k, \ \text{for} \ n = 1, 2, 3, \ldots, \] (1.2)

where $\hbar = h/2\pi$ is the reduced Planck constant or Dirac constant, while $n$ is the average phonon occupation number that satisfies Bose-Einstein statistics $f_{BE} = \frac{1}{\exp(\hbar \omega/k_B T) - 1}$.

Unlike the assigned energy given by Eq. (1.2), phonons themselves do not physically carry momentum. As an analogy to photons and matter waves, phonons can be treated with wave vector $k$, and thus assigned a pseudomomentum $\hbar k$ to distinguish from the real physical momentum.

In addition to the characteristics such as the angular frequency, or wavevector, polarization is another important feature of phonons. There are two modes of polarization for phonons: longitudinal mode and transverse mode. In the case of longitudinal modes, the displacement direction of the atoms from their equilibrium position coincides with the propagating direction of the wave, whereas for transverse modes, atoms move perpendicularly to the propagating direction of the wave. In a single chain of atoms, which can be equivalently treated as many rigid balls connected by Hooke’s law springs, the lattice wave is solely longitudinal. However, in three-dimensional (3D) situations (a cube that consists of many atoms), there exist three possible polarization modes: one longitudinal mode as we observed in the single chain, and two transverse modes.

Furthermore, two phonon branches will appear if we consider more than one atom per unit cell, which is always the case. The difference between acoustic branch and optical branch is illustrated in Fig. 1.3. In optical branch, the two neighboring atoms are moving out of phase, whereas in acoustic branch, they are moving in phase. Accordingly, assuming that there are $N$ atoms per unit cell, 3 acoustic branches and $3N-3$ optical branches will show up. Among them, the propagation speeds of optical phonons are almost zero, therefore optical phonons usually do not contribute to heat transport. The acoustic branch is mainly responsible for energy transport. Their propagation speeds, or wavevectors $k$, vary according to their energy $\hbar \omega$, or frequency $\omega$. In lattice dynamics, the relation between frequency and wavevector $\omega(k)$ is denoted as phonon dispersion.

1.2.2 Phonon dispersion

For simplicity, here we consider a linear chain of two different atoms with mass $m_1$ and $m_2$, respectively, as shown in Fig. 1.3. The offset distance between the equilibrium positions of two neighboring atoms is $a$ (the lattice constant). Define $u_j$ as the displacement of $j^{th}$ atom, so that the force on $(2j)^{th}$ and $(2j + 1)^{th}$ atom from its neighbors can be written as

\[ F_{2j} = K \left( u_{2j+1} - 2u_{2j} + u_{2j-1} \right), \]
\[ F_{2j+1} = K \left( u_{2j+2} - 2u_{2j+1} + u_{2j} \right), \] (1.3)
Figure 1.3: Schematic of optical and acoustic branches of phonons. The top atoms chain shows the equilibrium state of a 1D lattice. The middle one illustrates the optical phonon branch, where two neighboring atoms are always moving in phase. The bottom one denotes the acoustic phonon branch where their movements are out of phase. Picture by Brews Ohare

where $K$ is the spring constant. According to dynamics, the equation of motion can be written as

$$m_1 \frac{\partial^2 u_{2j}}{\partial t^2} = F_{2j} = K\left(u_{2j+1} - 2u_{2j} + u_{2j-1}\right),$$

$$m_2 \frac{\partial^2 u_{2j+1}}{\partial t^2} = F_{2j+1} = K\left(u_{2j+2} - 2u_{2j+1} + u_{2j}\right).$$

(1.4)

A trial solution set is $u_{2j} = A \exp\left[i (2jka \pm \omega t)\right]$ and $u_{2j+1} = B \exp\left[i ((2j + 1)ka \pm \omega t)\right]$. Substituting this solution set into Eq. (1.4) yields the dispersion

$$\omega^2 = K \left( \frac{1}{m_1} + \frac{1}{m_2} \right) \pm K \sqrt{\left( \frac{1}{m_1} + \frac{1}{m_2} \right)^2 - 4 \left( \frac{\sin^2 ka}{m_1 m_2} \right)}. \quad (1.5)$$

This is only an ideal case to illustrate phonon dispersion. A practical phonon dispersion in typical dielectric materials, silicon for instance, is similar in trend but embellished by a number of detailed modifications, as given in Fig. 1.5.

Thus, it can be seen that phonon propagation depends on phonon polarization, frequency and branch. Phonons will travel without damping due to their wave nature if there is no energy dissipating event happens, which equivalently implies their infinite lifetime. However, the lifetime of phonon is finite because the energy dissipating events do physically exist. These events are termed as phonon scattering as a whole.

1.2.3 Phonon scattering

Physically, there are several distinct mechanisms that cause phonons to scatter as they travel through the material: phonon-phonon scattering, phonon-impurity (mass-difference) scattering, phonon-electron scattering, and phonon-boundary scattering.

**Phonon-phonon scattering** As suggested by its name, phonon-phonon scattering describes the collisions among phonons. However, due to the wave nature of phonon, the overall pseudo-momentum of phonons is not necessarily conserved. In the cases of three phonon scattering,
Figure 1.4: The dispersion of optical and acoustic phonon in first Brillouin zone. Acoustic branch dispersion is given by taking the minus sign in Eq. (1.5), and optical branch dispersion is obtained by taking the plus sign in Eq. (1.5).

Figure 1.5: Phonon dispersion in silicon for both acoustic and optical phonons [4].
the momentum relation in a momentum-conserved situation can be written as

\[ k_1 + k_2 - k_3 = 0, \]  

(1.6)

where \( k_j \) with \( j = 1, 2, 3 \) is the wavevector for numbered phonons involved in the scattering event. The process formulated by Eq. (1.6) is named normal process (N process). Such a process does not result in any resistance for phonon propagation but merely redirects it. Thus this process is also classified as “non-resistive” process. According to McNelly et al. [5], this process provides major contribution to the appearance of “second sound” in thermal wave experiments at low temperature, which will be studied in Chapter 5. On the other hand, in the momentum-destroying scattering termed as Umklapp process (U process), the momentum relation becomes

\[ k_1 + k_2 - k_3 = G, \]  

(1.7)

where the additional \( G \) is modulo, a reciprocal lattice vector [6], leading to the resistance for phonon propagation. More details regarding the link between these two processes are unfortunately not well-understood. For example, Maznev et al. [6] pointed out that, an infinitesimal disturbance on wavevector in a given primitive cell may turn a N process to a U process.

**Mass-difference scattering** Mass-difference scattering is the scattering of phonons due to the mass difference between the crystal atoms and impurity atoms. The relaxation time of this scattering process is related to the concentration of impurities, the mass of impurity atoms and the group velocity of the phonon scattered.

**Phonon-electron scattering** Phonon-electron scattering refers to the scattering between phonons and electrons. The collisions between fast-moving electrons and the lattices may introduce significant energy exchange between the electrons and phonons, meanwhile bringing in changes of phonon characteristics depending on the concentration of electrons and many other conditions.

**Phonon-boundary scattering** This scattering process happens when the phonons hit the boundary, where the symmetry is broken due to the termination of bulk crystal structure. In total, three options are available for these phonons: to transmit if possible, to be reflected or to become a surface phonon that propagates along the surface and decays into the depth of the material. Phonon-boundary scattering can be semi-empirically formulated depending on if the scattering is diffuse or specular. Casimir introduced a well-known Casimir limit [7] for diffuse scattering.

Under relaxation time approximation (BGK approximation), which will be introduced later, each of these mechanisms can be characterized by a relaxation time \( \tau \). According to Matthiessen’s rule, there defines a combined relaxation time to account for an overall strength of scattering processes:

\[ \frac{1}{\tau} = \frac{1}{\tau_U} + \frac{1}{\tau_M} + \frac{1}{\tau_{ph-b}} + \frac{1}{\tau_{ph-e}}, \]  

(1.8)
where in the subscript, $U$, $M$, ph-b and ph-e denote Umklapp, mass-difference, phonon-boundary and phonon-electron scattering, respectively. Thus, the combined relaxation time leads to the simplification of real situations by skipping the detailed features for different scattering mechanisms. But such an average relaxation time may fail if some scattering mechanisms are dominant so that the phonon transport performs their unique features.

With so many phonon characteristics, the simulation of phonon transport is unaffordably expensive in microscale, if all the detailed physics are considered. Furthermore, the technical difficulties in experiments also hinder the direct observation of phonon transport. Although we do have reasonably good approximation in macroscopy, the Fourier’s law, certain deviations recently have been found frequently between the predictions by Fourier’s law and the experimental results, especially when the system length scale is approaching micrometer or even nanometer scale.

1.3 Current state of the art: mesoscale models

In the ballistic transport regime, where the length scale is close to a nanometer at room temperature, Molecular Dynamics (MD) simulations offer reasonable solutions by numerically simulating the motion of the atoms or molecules; while in diffusive regime, Fourier’s law holds. Between these two extreme cases called mesoscale, for example Kn ~ 1, the computational expense for MD is very high, and Fourier’s law breaks down due to the invalidity of diffusive assumption. New models are necessary. Unfortunately, numerous devices fall right into this regime, raising the demands for the development of models for this regime called the mesoscale regime.

1.3.1 Theory of Boltzmann transport equation

In the mesoscale, the Boltzmann transport equation (BTE), devised by Ludwig Boltzmann, offers a description on the statistical behavior of particles, and can be implemented in phonon transport. It is commonly used in situations where the system’s state is far from thermodynamic equilibrium.

In the BTE, the distribution function of the particles is defined so as to capture their statistical behavior. The distribution function $f(t, r, p)$ represents the density of the particles at position $r$ with momenta $p$. Considering an external force $\mathbf{F} = dp/dt$ on the particles of mass $m$ and the scattering between the particles, we have [8]

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla_r f + \frac{\mathbf{F}}{m} \cdot \nabla_v f = \left( \frac{\partial f}{\partial t} \right)_{\text{scattering}}.$$  \hfill (1.9)

For phonon, we have momenta $p = mv$, and wavevector $k = \hbar v$. In wavevector form, Eq. (1.9) can be written as

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla_r f + \frac{\mathbf{F}}{\hbar} \cdot \nabla_k f = \left( \frac{\partial f}{\partial t} \right)_{\text{scattering}}.$$  \hfill (1.10)

Based on the distribution function solved by the BTE, a volume average physical quantity in macroscale
can be linked to the physical quantity of particles by [9]

\[
\langle X(\mathbf{r}) \rangle = \frac{1}{V} \sum_{\mathbf{k},s} X(\mathbf{r}, \mathbf{k}) f = \frac{1}{(2\pi)^3} \int X(\mathbf{r}, \mathbf{k}) f \, d^3k,
\]  

(1.11)

where \(d^3k = dk_x dk_y dk_z\). However, the scattering term introduces the main challenge for the BTE. As mentioned in the previous section, several distinct scattering mechanisms simultaneously contribute to this term, adding complexity to the mathematical formulation of the BTE. To illustrate the complexity, we only show the final formula of the scattering between two particles. A detailed and rigorous derivation can be found in Ref. [8]. Assume that initially the wavevector and distribution function for these two particles are \(\mathbf{k}_1, \mathbf{k}_2\) and \(f(t, \mathbf{r}, \mathbf{k}_1), f(t, \mathbf{r}, \mathbf{k}_2)\), while after scattering they become \(\mathbf{k}_1', \mathbf{k}_2'\) and \(f(t, \mathbf{r}, \mathbf{k}_1'), f(t, \mathbf{r}, \mathbf{k}_2')\). Accordingly, the mathematical formula of this scattering can be expressed as

\[
\left( \frac{\partial f}{\partial t} \right)_{\text{scattering}} = K' \int W \times [f(t, \mathbf{r}, \mathbf{k}_1') f(t, \mathbf{r}, \mathbf{k}_2') - f(t, \mathbf{r}, \mathbf{k}_1) f(t, \mathbf{r}, \mathbf{k}_2)] 
\times \frac{d^3k_2 d^3k_1'}{d^3k_2' d^3k_1} d^3k_2 d^3k_1 d^3k_2',
\]  

(1.12)

where \(K' = \frac{V^3}{(2\pi)^9}\) is a conversion factor, and \(W\) is the corresponding quantum state transition probability [8]. Replacing the right hand side (RHS) of Eq. (1.10) with Eq. (1.12) finally yields

\[
\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{r} f + \frac{\mathbf{F}}{\hbar} \cdot \nabla \mathbf{k} f = K' \int W \times [f(t, \mathbf{r}, \mathbf{k}_1') f(t, \mathbf{r}, \mathbf{k}_2') - f(t, \mathbf{r}, \mathbf{k}_1) f(t, \mathbf{r}, \mathbf{k}_2)] 
\times \frac{d^3k_2 d^3k_1'}{d^3k_2' d^3k_1} d^3k_2 d^3k_1 d^3k_2'.
\]  

(1.13)

After applying scattering terms, the BTE ends up with a complicated integro-differential equation. Similar to the radiative transport equation (RTE), it can be inferred that large computational costs have to be consumed if the BTE is directly solved, leaving aside the unknown details for distinct scattering mechanisms. From engineering perspective, both the computational costs and the unknown details of scattering make this solution far from applicable. For this reason, simplified BTE models are developed to lower the computational costs and provide guidance for engineering applications and experimental data analysis.

### 1.3.2 Current simplified models

One simplification for the scattering processes should be noted before introducing simplified models compatible with BTE, that is the BGK approximation, or relaxation time approximation. It is the joint work of Bhatnagar, Gross and Krook [10], stating that the scattering process can be approximated by a relaxation process with a characteristic time \(\tau\):

\[
\left( \frac{\partial f}{\partial t} \right)_{\text{scattering}} = -\frac{f - f^0}{\tau},
\]  

(1.14)

where \(f^0\) is the distribution function at equilibrium. For phonons, this is Bose-Einstein distribution, \(f^0 = f_{BE}\). Different scattering categories can all be treated with corresponding relaxation times as previously mentioned in phonon scattering section. If Matthiessen’s rule of Eq. (1.8) is applied, we
rewrite BTE without external force \((F = 0)\) as

\[
\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla_{\text{r}} f = -\frac{f - f^0}{\tau}.
\]  

(1.15)

One most simplified case is that, only one frequency of phonon is considered so that the phonon group velocity is a constant. This simplified BTE is commonly used and known as gray BTE, due to the independence on frequency, or color-blindness. It equivalently introduces a linear dispersion of phonon. Based on this approximation, several constitutive nondiffusive models derivable from BTE are introduced here.

**C-V model** This model was proposed by Cattaneo [11] and Vernotte [12] initially to remove the paradox of infinite heat propagation speed from Fourier’s law. This model is also known as hyperbolic heat conduction equation, introducing a finite propagating speed for the heat wave. Mathematically, this equation can be written as

\[
\tau \frac{\partial q}{\partial t} + q = -\kappa \nabla T,
\]  

(1.16)

where \(\tau\) is the relaxation time or build up period for the commencement of heat flow with an imposed temperature gradient. The relaxation \(\tau\) is an extension of the relaxation time in BTE for rarefied gases to solids case, and thus arguably introduces the wave nature. However, some researchers found a violation of second thermodynamic law in this model [13], which is the most serious criticism on this model.

**G-K model** This model was proposed by Guyer and Krumhansl [14] in 1966, initially for explaining the second sound discovered in thermal wave experiments. Mathematically, this model can be expressed as

\[
\tau \frac{\partial q}{\partial t} + q = -\kappa \nabla T + \Lambda^2 \left( \nabla^2 q + 2 \nabla \nabla \cdot q \right),
\]  

(1.17)

where \(\Lambda\) is the phonon MFP. This model shows better performances on elevating the fast variables, such as the heat flux \(q\), to the status of independent variables at the same level as the slow variables, such as temperature [15].

**EPRT** The equation of phonon radiative transfer (EPRT) is described by Majumdar [16] to connect radiation theory to phonon transport in dielectric materials. Analogous to the intensity of light, it defines the intensity of phonon \(I\) as

\[
I(\theta, \phi, \omega, x, t) = \sum_p v(\theta, \phi) f(x, t) \hbar \omega D(\omega),
\]  

(1.18)

where \(p\) is for polarizations, \(\theta\) and \(\phi\) are polar and azimuthal angle, respectively, \(\omega\) is the phonon frequency, and \(D(\omega)\) is the density of state of phonon with frequency \(\omega\). This is a 1D example due to the brevity consideration. With Eq. (1.18), BTE can be rewritten as

\[
\frac{\partial I}{\partial t} + v_x \frac{\partial I}{\partial x} = -\frac{I - I^0}{\tau},
\]  

(1.19)
\( v_x = v \cos \theta \) is the projection of real velocity in \( x \) direction. The temperature and heat flux can accordingly be determined with solved intensity \( I \). However, this analogy between phonon and photon is not always true, especially when phonon-phonon scattering is taking an important part of the transport process. By the definition of intensity, the phase of lattice waves is lost, which is one characteristic of phonons related to their interaction.

**Ballistic-diffusive equation** This model is proposed by Chen [17]. The main idea for this model is to categorize phonons into two groups, ballistic phonons and diffusive phonons, and accordingly split the distribution function and heat flux into two parts: \( f = f_b + f_d \), \( \mathbf{q} = \mathbf{q}_b + \mathbf{q}_d \). Starting from the BTE, we here skip the derivation and directly deliver the governing equation

\[
\tau_d \frac{\partial \mathbf{q}_d}{\partial t} + \mathbf{q}_d = -\kappa \nabla T_d, \text{ for diffusive phonons (1.20)}
\]

\[
\tau_b \frac{\partial \mathbf{q}_b}{\partial t} + \mathbf{q}_b = -\kappa \nabla T_b + \Lambda_b^2 \left( \nabla^2 \mathbf{q}_b + 2 \nabla \nabla \cdot \mathbf{q}_b \right), \text{ for ballistic phonons (1.21)}
\]

where \( \Lambda_b \) is the MFP of the ballistic phonon. Mathematically, this equation form is pretty similar to a combination of G-K model and C-V model.

There are many other nondiffusive models, like C- and F-process model [18], a dual-phase lag (DPL) model [19], a two-channel model [20]. These models can be solved combined with energy conservation given as

\[
c_v \frac{\partial T}{\partial t} + \nabla \cdot \mathbf{q} = Q, \quad (1.22)
\]

where \( c_v \) is the volumetric specific heat, and \( Q \) is the volumetric energy generation rate. Unfortunately, there still lacks a well-recognized model widely supported by different experiments and is applicable for engineering use. Currently, Fourier’s law with effective thermal conductivity is still in use for experimental data analysis, which is by nature questionable.

### 1.4 Remaining challenges & objectives

As described in this chapter, we can see that Fourier’s law gives inaccurate predictions and is not applicable in mesoscale. On the other hand, tools such as MD for nanoscale heat conduction are too expensive for in the mesoscale. Consequently, a model designed specifically for mesoscale heat conduction is a desirable choice. Once found, this model can benefit both engineering applications and fundamental study.

At present, there exists no low-cost mesoscale heat conduction model, or nondiffusive model, that bridges the gap between MD and Fourier’s law for either engineering use or fundamental study. However, recently, two compatible nondiffusive heat transfer models were proposed. One is the Two-Parameter Heat Conduction (TPHC) model proposed by Ma [21], the other is the Enhanced Gray Boltzmann transport equation proposed by Wang [22]. Although one unified model that satisfies the requirements from both engineering applications and fundamental study is not found, these two models together show potential to provide satisfactory solutions. In this thesis, we want to provide solutions for this main challenge based on these two models.

Accordingly, we briefly list the objectives from these two perspectives.
1. Offer a reliable nondiffusive solution for engineering applications that satisfies the following requirements
   (a) It is able to predict nondiffusive heat transfer with satisfactory precision according to project requirements.
   (b) It is mathematically simple and computationally affordable.
   (c) It offers a criterion that serves as a guidance to decide if nondiffusive models must be applied before the simulation is carried out.

2. Offer a reliable nondiffusive solution for fundamental study that satisfies the following requirements
   (a) It is able to bridge the gap between MD and Fourier’s law.
   (b) It provides new findings and possible guidance for experimental explorations.
   (c) It is appropriately simplified to lower the computational costs.

1.5 Outline

- In Chapter 2, the nondiffusive heat transfer models, the Two-Parameter Heat Conduction model and Enhanced Gray Boltzmann transport equation, that satisfy our stated requirements are developed, and the physical interpretation of the parameters in the models is offered.
- In Chapter 3, the numerical algorithms for solving the Two-Parameter Heat Conduction model and Enhanced Gray Boltzmann transport equation are described.
- In Chapter 4, the Two-Parameter Heat Conduction model is validated both analytically and numerically by comparing to multiple nondiffusive experiments, preparing it for engineering projects.
- In Chapter 5, the Enhanced Gray Boltzmann transport equation is validated both analytically and numerically by experiments in different heat conduction regimes, revealing the hidden physics in mesoscale and providing hints for further study.
- In Chapter 6, a summary for this thesis is given and future work is suggested.
Chapter 2

Nondiffusive Heat Transfer Models

In this chapter, a constitutive Two-Parameter Heat Conduction (TPHC) model is introduced at the beginning with a nondimensional parameter defined to characterize the nondiffusive effects, serving as a criterion to demarcate the limit up to which Fourier’s law is still applicable. Then the derivation of an Enhanced Gray BTE (EG-BTE) for the phonon transport in dielectric materials is presented. Next, the compatibility between these two models is shown, which explains the similarity between these two models.

2.1 Two-Parameter Heat Conduction Model

In this section, we introduce the Two-Parameter Heat Conduction (TPHC) model proposed by Ma [21]. We derive the TPHC model based on the definition of nonequilibrium absolute temperature $\beta$ in a one-dimensional (1D) case.

2.1.1 Derivation of the TPHC model from the definition of $\beta$

We start with the equilibrium status of the system. Under the framework of diffusive heat transfer, there exists a local thermodynamic equilibrium temperature $T$. In this diffusive regime, the relation between the thermodynamic equilibrium temperature $T$ and heat flux $q$ satisfies Fourier’s law. Together with energy conservation in 1D, we have

$$\frac{\partial E}{\partial t} + \frac{\partial q}{\partial x} = Q, \quad (2.1)$$

$$q = -\kappa \frac{\partial T}{\partial x}, \quad (2.2)$$

where $\kappa$ is the thermal conductivity, and $Q$ is the volumetric heat generation rate.

However, the thermodynamic equilibrium may not be fully established or even far away from established in the regime of nondiffusive heat transfer. According to Cimmelli et al. [23], a nonequilibrium absolute temperature $\beta$ can be defined as $\beta = E/c_v$ with the volumetric energy density $E$. $c_v$ here is the volumetric specific heat. It remains roughly a constant if the temperature change is not very big (i.e., tens Kelvin), so that $\frac{\partial E}{\partial t} = c_v \frac{\partial \beta}{\partial t}$ holds. With this relation, Eq. (2.1) can thus be written
as
\[ c_v \frac{\partial \beta}{\partial t} + \frac{\partial q}{\partial x} = Q, \quad (2.3) \]

The nonequilibrium absolute temperature \( \beta \) can be linked to \( T \) through a relaxation process
\[ \frac{\partial \beta}{\partial t} = -\frac{\beta - T}{\tau_B}, \quad (2.4) \]

where \( \tau_B \) is the relaxation time for this process. Rewrite this equation
\[ T = \beta + \tau_B \frac{\partial \beta}{\partial t}, \quad (2.5) \]

and substitute Eq. (2.5) into Eq. (2.2) to replace \( T \). Assuming there is no external heat source \( (Q = 0) \), together with Eq. (2.3), we have
\[ c_v \frac{\partial \beta}{\partial t} + \frac{\partial q}{\partial x} = 0, \quad (2.6) \]
\[ q = -\kappa \frac{\partial \beta}{\partial x} + \kappa \tau_B \frac{\partial^2 q}{\partial x^2}, \quad (2.7) \]

According to kinetic theory, the thermal conductivity \( \kappa \) can be linked to the volumetric specific heat \( c_v \), speed of sound \( c \), and diffusive phonon lifetime \( \tau_D \) [21] through
\[ \kappa = c_v c^2 \tau_D / 3. \quad (2.8) \]

If the diffusive phonon mean free path (MFP) is defined by \( \Lambda_D = c \tau_D \), similarly, the ballistic phonon MFP can be defined by \( \Lambda_B = c \tau_B \). Plugging these two parameters into Eq. (2.7) gives 1D TPHC model:
\[ q = -\kappa \frac{\partial \beta}{\partial x} + \frac{\Lambda_D \Lambda_B}{3} \frac{\partial^2 q}{\partial x^2}. \quad (2.9) \]

In this equation, there are two independent parameters \( (\kappa \text{ and } \Lambda_B) \) that will be determined from the experiments. Because of these two independent parameters, this model is called Two-Parameter Heat Conduction model.

According to Ma [21], together with the conservation of energy in three-dimensional form, the 3D TPHC model is given as
\[ c_v \frac{\partial \beta}{\partial t} + \nabla \cdot q = Q, \quad (2.10) \]
\[ q = -\kappa \nabla \beta + \frac{\Lambda_D \Lambda_B}{9} \left( \nabla^2 q + 2 \nabla (\nabla \cdot q) \right). \quad (2.11) \]

If appropriate initial and boundary conditions are given, the TPHC model can be solved analytically or numerically using a standard numerical algorithm, for example, finite difference method. Here we briefly introduce the implementation of initial and boundary condition.

The initial conditions in heat transfer problems are usually given as an initial temperature
field. Since the TPHC model is directly linked to temperature, this initial temperature field can be directly plugged in for calculation. On the other hand, the boundary condition in heat transfer problem are usually given as boundary temperature or boundary heat flux, or a mixture of these two (i.e., convection). Either in the case with boundary temperature specified, or in the case with boundary heat flux, we can directly plug it into the computation. For a case with a mixed type, it can be decomposed into the previous two.

From Eq. (2.11), it can be seen that the TPHC model is reducible to Fourier’s law once $\Lambda_B = 0$. On the other hand, if $\Lambda_B$ is adequately large, the nondiffusive effects contributed by the last two terms on the right hand side (RHS) of Eq. (2.11) weigh more in the heat transfer. Therefore, based on this TPHC model, a nondimensional parameter that characterizes the weighs from the nondiffusive effects can be defined, as shown in the following subsection.

### 2.1.2 Characterizing nondiffusive heat transfer using the TPHC model

Although we already provide the TPHC model for nondiffusive heat transfer, Fourier’s law is still preferred in engineering projects. This is because it requires lower computational costs compared to nondiffusive models, and is more convenient to apply (it has already been integrated into many commercial softwares). However, as introduced in Chapter 1, Fourier’s law may fail under certain circumstances. A lack of criterion to evaluate its applicability in the given situation results in possible defects in the products. For example, one chip is designed according to Fourier’s law, but the typical size of the transistors is so small that Fourier’s law actually fails here. This failure leads to the decrease in thermal conductivity, and thus the heat removal does not satisfy the requirements, and thus the chip may break down due to thermal failure. On the other hand, a simulation that produces identical results as Fourier’s law but consumes much more time is by no means desirable. These two simple examples illustrate the importance of the criterion for engineering projects. In this subsection, we define a nondimensional parameter based on the TPHC model that can be used as this criterion.

Specifically, the value of criterion $\zeta_c$ should be determined according to the project requirements. For example, typically, 1% can be considered as a small number in engineering projects. Assuming this, the criterion $\zeta_c$ can be set as 0.01. But it should be noted that $\zeta = 0.01$ can predicts neither 1% difference in temperature decay rate nor 1% difference in heat flux (these sayings
themselves are ambiguous). We will offer more illustrations and validations on this criterion in the applications of the TPHC model.

Next, we move on to the model for fundamental study, the Enhanced Gray Boltzmann transport equation.

### 2.2 Enhanced Gray Boltzmann Transport Equation

In dielectric materials, the main heat carrier for energy transport is phonons. As introduced in Chapter 1, a phonon is defined as quasiparticle to describe lattice vibration modes of elastic solid crystals in quantum mechanical view. It is essentially quantized energy and does not carry any physical momentum (only pseudo-momentum). The state of a phonon in phase space is characterized by a distribution function $f(t, r, v)$, where $t$ means time, $r$ represents a coordinate vector, and $v = v(\omega, p)$ is phonon group velocity vector related to phonon frequency $\omega$ and polarization $p$ (longitudinal acoustic (LA), transverse acoustic (TA), longitudinal optical (LO) and transverse optical (TO)). This distribution function changes to $f(t + \Delta t, r + \Delta r, v + \Delta v)$ at the time $t + \Delta t$. During this time interval $\Delta t$, the number of phonons may not be conserved due to the scattering processes, which denote the interactions among phonons or with impurities and boundaries. As a consequence, assuming the net rate of phonon generation is $\left(\frac{\partial f}{\partial t}\right)_{c}$, the change of distribution function can be written as

$$f(t + \Delta t, r + \Delta r, v + \Delta v) - f(t, r, v) = \left(\frac{\partial f}{\partial t}\right)_{c} \Delta t + Q \Delta t. \quad (2.13)$$

On the right hand side (RHS) of Eq. (2.13), $Q$ is the external source term, and $\left(\frac{\partial f}{\partial t}\right)_{c}$ denotes the change rate of phonon distribution function due to the scattering processes as they are traveling at group velocity $v = V_\omega$ in the media. The causes of scattering have been summarized in Chapter 1. Scattering due to different mechanisms can generally be described with relaxation time approximation with a characteristic relaxation time corresponding to each mechanism (BGK approximation). These relaxation times, for simplicity, can be approximated as one average relaxation time [24], which can be written as

$$\left(\frac{\partial f}{\partial t}\right)_{c} = -\frac{f - f^0}{\tau_R}, \quad (2.14)$$

where $\tau_R = \tau_R(\omega, p, T)$ is the relaxation time ($T$ is absolute temperature), and $f^0$ represents the local equilibrium distribution function. $\tau_R$, according to kinetic theory, can be linked to thermal conductivity $\kappa$ as $\tau_R = 3\kappa/c_v c^2$, where $c_v$ is the volumetric specific heat and $c$ is the speed of sound in the crystal. The local equilibrium distribution function for phonon is given by the Bose-Einstein distribution

$$f^0 = f_{BE} = \frac{1}{\exp\left(h\omega/k_BT\right) - 1}, \quad (2.15)$$

where $h$ is the reduced Planck constant (Planck constant $h$ divided by $2\pi$), $\omega$ is the phonon angular frequency, $k_B$ is Boltzmann constant, and $T$ is the absolute temperature.

It should be noted that the variables involved have dependence on frequency, polarization
and direction, which results in the complexity of this equation. To simplify the equation, one most used assumption is the gray-medium assumption, which helps to eliminate the phonon frequency dependence of group velocity and relaxation time. Moreover, only acoustic branch is considered while the optical branch is assumed to have negligible contribution to heat transfer. At room temperature, Debye velocity is usually taken as the sound speed. It is an average of two polarization modes \( 3c^{-3} = c_l^{-3} + 2c_t^{-3} \), where the subscript \( l \) means longitudinal mode and \( t \) means transverse mode. After these simplifications, the magnitude of phonon group velocity \( v \) (the speed of sound \( c \)) and relaxation time \( \tau_R \) are both constant.

Based on gray assumption and assuming there are no external forces, following the process proposed by Wang et al. [22], we truncate the Taylor expansion of the left hand side (LHS) of Eq. (2.13) up to the second order terms to obtain

\[
\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla f + \frac{\tau_c}{2} \left[ \frac{\partial^2 f}{\partial t^2} + 2\mathbf{v} \cdot \nabla \frac{\partial f}{\partial t} + (\mathbf{v} \cdot \nabla)^2 f \right] = -\frac{f - f^0}{\tau_R} + Q, \tag{2.16}
\]

where \( \tau_c \) is a characteristic time describing the nonlocal effects from ballistic phonon transport [22]. According to Eq. (4) by Wang et al. [22], the last term on the LHS does not contribute to heat transfer and thus can be omitted. In some cases, the second order time derivative term in Eq. (2.16) can be omitted depending on the time scale of interest, but in some other studies, as will be shown in Chapter 5, this term has significant impacts and has to be considered. Although the coefficient of this term is \( \frac{\tau_c^2}{\tau_R} \) according to Taylor expansion, it needs modification, as shown below.

Without loss of generality, we start with 1D EG-BTE with an undetermined constant \( a_c \) given by

\[
a_c \tau_c \frac{\partial^2 f}{\partial t^2} + \frac{\partial f}{\partial t} + \mu v \frac{\partial f}{\partial x} = -\frac{f - f^0}{\tau_R} - \mu \tau_c \frac{\partial^2 f}{\partial x \partial t} + Q, \tag{2.17}
\]

where \( \mu = \cos \theta \) is the directional cosine and \( \theta \) is the polar angle. According to Taylor expansion, \( a_c = \frac{1}{2} \). However, the gray assumption implemented here leads to this undetermined coefficient, \( a_c \), because this assumption presumes the linear dispersion relation of phonon, which may not be a good approximation here since the change of \( \tau_c \) and \( v \) is not always linear. To stay with the simplifications provided by the gray assumption, we determine this indefinite coefficient by analyzing the dispersion relation of Eq. (2.17).

Assuming \( e \) can be decomposed into the dynamic part \( f' \) and the equilibrium part \( f^0 \), \( f = f' + f^0 \), we then obtain an equation for \( f^0 \), which is directly related to temperature \( T \)

\[
a_c \tau_c \frac{\partial^2 f^0}{\partial t^2} + \frac{\partial f^0}{\partial t} + \mu v \frac{\partial f^0}{\partial x} = -\tau_c \mu \frac{\partial^2 f^0}{\partial t \partial x}. \tag{2.18}
\]

The dynamic component \( f' \) determines the energy flow, but it is usually the temperature, equivalently the equilibrium component \( f^0 \), that marks the signal. Therefore, we determine the coefficient \( a_c \) by analyzing Eq. (2.18).

By introducing a plane-wave solution \( f^0 = \exp \{i(kx - \omega t)\} \) to Eq. (2.18), we obtain the dispersion relation for Eq. (2.18),

\[
k = \frac{a_c \tau_R \tau_c \omega + iT \sqrt{\omega}}{\tau_R \tau_c \omega + IT \mu}, \tag{2.19}
\]
where $k$ is a complex wavenumber, and $\omega$ is the angular frequency represents the wave frequency. This dispersion should be distinguished from the dispersion of phonons. Accordingly, the phase velocity $v_p$ of the wave can be determined as

$$v_p = \frac{1 + R^2 \omega^2}{1 + aR^2 \omega^2 \mu v},$$

(2.20)

where $R$ is the ratio defined as $R = \tau_c / \tau_R$. $v_p$ here should never exceeds the first sound speed $c$. Recall that the coefficient by Taylor expansion is $a_c = 1 / 2$. The dispersion of thermal wave with $a_c = 1 / 2$ and $R = 1$ is given in Fig. 2.1 as an example.

It can be found through this study that there discovers a thermal wave with even higher speed ($1 / a_c$ times) than the first sound if $a_c < 1$. The omission of this term is a special case where $a_c = 0$ and phase velocity $v_p \rightarrow \infty$ for high $\omega$ range (initial stage of the thermal transport). Consequently, to maintain the physics of sound wave, the coefficient is determined as $a_c = 1 / 2$ rather than $a_c = 0$. Substituting $a_c = 1$ into Eq. (2.17), a constant phase velocity that corresponds to the speed of sound persists, which is compatible with gray medium assumption. This improvement can be seen when the thermal transport time scale is small, i.e., close to $\tau_c$. Thus, we extend the applicability of the EG-BTE to a wider time scale by considering the second order time derivative. The EG-BTE can be written as

$$\tau_c \frac{\partial^2 f}{\partial t^2} + \frac{\partial f}{\partial t} + \mu v \frac{\partial f}{\partial t} = - \frac{f - f^0}{\tau_R} - \mu v \tau_c \frac{\partial^2 f}{\partial x \partial t} + Q.$$  \tag{2.21}

Eq. (2.21) is the EG-BTE in distribution function form. In gray medium, the distribution function $f$ can be related to the equilibrium distribution function $f^0$ by

$$f^0 = \frac{1}{4\pi} \int_{4\pi} f d\Omega.$$  \tag{2.22}

To establish a direct link between the distribution function and temperature, we follow the same process described by Wang et al. [22] to obtain EG-BTE in energy form [25, 26] by defining

$$e_\omega = \frac{1}{4\pi} \hbar \omega D(\omega), \quad e = \int_0^{\omega_D} e_\omega d\omega, \quad E = \int_4^{4\pi} e d\Omega,$$

(2.23)

where $e_\omega$ is the volumetric energy density per unit frequency and solid angle, $e$ is the volumetric energy density per solid angle, and $E$ is the volumetric energy density. $\Omega$ here represents the solid angle, $D(\omega)$ represents the phonon density of states, and $\omega_D$ is Debye cutoff frequency. According, we have

$$e^0 = \frac{1}{4\pi} \int_{4\pi} e d\Omega.$$  \tag{2.24}

Thus, we obtain the energy form of EG-BTE by multiplying both sides of Eq. (2.21) with $\hbar \omega D(\omega) / 4\pi$ and integrating over all frequency range:

$$\tau_c \frac{\partial^2 e}{\partial t^2} + \frac{\partial e}{\partial t} + \nabla e = - \frac{e - e^0}{\tau_R} - \tau_c v \cdot \nabla \frac{\partial e}{\partial t} + Q.$$  \tag{2.25}
Figure 2.1: Dispersion of Eq. (2.17) with $a_c = \frac{1}{2}$ as an example. (a). Phase velocity; (b). Imaginary part of the wavenumber. The phase velocity reaches twice of the first sound speed.

Eq. (2.25) is the EG-BTE.

$e^0$ here corresponds to the equilibrium distribution function $f^0$ and is accordingly the equilibrium energy density per unit solid angle. $e^0$ can be related to temperature as

$$e^0 = \frac{1}{4\pi} \int_0^{\omega_D} h\omega f^0(T)D(\omega) d\omega \approx \frac{1}{4\pi} c_v T,$$

(2.26)

where $c_v$ is the volumetric specific heat and $T$ is the temperature. Similarly, we also can relate $e$ to the heat flux [25] as

$$q = \int_{4\pi} \mathbf{v}ed\Omega = \int_{4\pi} \int_0^{\omega_D} \mathbf{v}h\omega f\frac{D(\omega)}{4\pi} d\omega d\Omega.$$  

(2.27)

Therefore, EG-BTE in Eq. (2.25) is applicable for calculating temperature and heat flux once appropriate boundary and initial conditions are specified.

The initial conditions are usually given as an initial temperature field at equilibrium so that $e^0$ can be calculated by Eq. (2.26), and thus $e$ can be determined accordingly through Eq. (2.24) assuming that $e$ is isotropic. On the other hand, typically, the boundary conditions in heat transfer problems are given as boundary temperature or boundary heat flux, or a mixture of these two.

**Given boundary temperature** In this case, the boundary temperature is specified as $T_w$. Similar
to the initial conditions, through Eqs. (2.26) and (2.24), $e$ can also be determined with the assumption that the wall is diffuse, or in other words, $e$ is isotropic.

**Given boundary heat flux** In this case, the heat flux at the boundary is specified as $q_w$. Equation (2.27) can be applied to relate $q_w$ to $e$ if the direction-dependence of $e$ can be appropriately treated. To take care of the direction-dependence, one option is to assume an isotropic $e$ so that the integral with respect to solid angle can be calculated according to the boundary geometry. The other option, usually applied on insulation, is to assume the specular reflection, so that the direction-dependence for boundary $e$ can be determined by the incident part of $e$.

**Mixed boundary** In this case, the relation between the boundary temperature and boundary heat flux is specified. This boundary is a combination of the two mentioned above, so that it can be decomposed and dealt following the processed for the two kinds of boundary conditions.

In addition, we will also illustrate the implementation of boundary conditions in Chapter 3, where the numerical schemes for solving EG-BTE will also be introduced.

Compared to the TPHC model with a nondimensional parameter, one may wonder if similar nondimensional parameter can also be defined based on the EG-BTE, which is more general compared with the TPHC model. Unfortunately, the special case with core parameter $\tau_c = 0$ for EG-BTE is gray BTE, and gray BTE is only comparable to Fourier's law if appropriate assumptions are specified. Hence, the nondimensional parameter $\zeta$ based on EG-BTE is not defined. When $\zeta$ is referred in this work, it is unambiguously based on the TPHC model.

In the next section, we will derive the TPHC model from EG-BTE to show their compatibility.

### 2.3 Compatibility between the TPHC model and EG-BTE

The compatibility between the TPHC model and EG-BTE lies in the derivability of the TPHC model from EG-BTE. In addition to starting from the definition of nonequilibrium absolute temperature $\beta$, the TPHC model can also be derived from the EG-BTE [9]. The key assumption for this derivation is [8, 16]

$$\nabla f = \frac{\partial f^0}{\partial T} \nabla T. \tag{2.28}$$

Under this assumption, we start from Eq. (2.25) and derive it term by term. First, we omit the first term on the LHS of Eq. (2.25) because whenever this term is important to heat transfer, the key assumption $\nabla f = \frac{\partial f^0}{\partial T} \nabla T$ already fails. Consequently, within the range of derivability of the TPHC model from EG-BTE, this term is not necessary to be considered.

Then, multiplying the second term $\frac{\partial f}{\partial t}$ with $v h \omega D(\omega) / 4\pi$ and integrating over frequency and solid angle yields

$$\int_{4\pi} \int_0^{\omega_0} v h \omega D(\omega) \frac{\partial f}{\partial t} d\omega d\Omega = \frac{\partial}{\partial t} \int_{4\pi} \int_0^{\omega_0} v h \omega f D(\omega) d\omega d\Omega = \frac{\partial q}{\partial t}, \tag{2.29}$$

where we assume that the sequence of the integration and the differentiation is exchangeable, and Eq. (2.27) has already been plugged in to convert distribution function $f$ to physical quantity $q$. 

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Similarly, multiplying the third term \( v \cdot \nabla f \) by \( \nu_0 D(\omega) / 4\pi \) and integrating over frequency and solid angle yields
\[
\int_{4\pi} \int_0^{\omega_0} v \omega \cdot \nabla f \frac{D(\omega)}{4\pi} d\omega d\Omega = \int_{4\pi} \int_0^{\omega_0} \nu_0 \omega \cdot \nabla f \frac{\partial f^0}{\partial T} \frac{D(\omega)}{4\pi} d\omega d\Omega
\]
\[
= \nabla T \int_0^{\omega_0} \frac{\nu_0^2}{3} \hbar \omega \frac{\partial f^0}{\partial T} D(\omega) d\omega = \frac{k}{\tau_R} \nabla T. \tag{2.30}
\]
In Eq. (2.30), we also assume that the integration and the gradient can be switched. Temperature \( T \) depends on neither angular frequency nor solid angle so it can be taken out of the integration. Moreover, according to Wang [9], given the thermal conductivity \( \kappa = \int_0^{\omega_0} \frac{\nu_0^2}{3} \hbar \omega \frac{\partial f^0}{\partial T} D(\omega) d\omega \), we can simplify this equation as shown above.

On the RHS of EG-BTE by Eq. (2.25), the first term \( \frac{\nu_0 f^0}{\tau_R} \) can similarly be written as
\[
\frac{1}{\tau_R} \int_{4\pi} \int_0^{\omega_0} v \omega (f - f^0) \frac{D(\omega)}{4\pi} d\omega d\Omega = \frac{q}{\tau_R}. \tag{2.31}
\]
In Eq. (2.31), both \( f^0 \) and \( v \) have no direction-dependence so that the integral of \( f^0 \) over all solid angle is zero, and the rest part can be linked to \( q \) according to Eq. (2.27).

At last, we can also rewrite \( \tau_c v \cdot \nabla \frac{\partial f}{\partial t} \) to be
\[
\tau_c \int_{4\pi} \int_0^{\omega_0} v \omega v \cdot \nabla \frac{\partial f}{\partial t} \frac{D(\omega)}{4\pi} d\omega d\Omega = \tau_c \nabla \frac{\partial T}{\partial t} \int_0^{\omega_0} \frac{\nu_0^2}{3} \hbar \omega \frac{\partial f^0}{\partial T} D(\omega) d\omega = \frac{\tau_c k}{\tau_R} \nabla \frac{\partial T}{\partial t}, \tag{2.32}
\]
Thus, after this term by term derivation, Eq. (2.25) can be written as
\[
0 + \frac{\partial q}{\partial t} + \frac{k}{\tau_R} \nabla T = -\frac{q}{\tau_R} - \frac{\tau_c k}{\tau_R} \nabla \frac{\partial T}{\partial t}. \tag{2.33}
\]
With the energy conservation given by Eq. (2.10), we can replace the last term on the RHS of Eq. (2.33) and rewrite it to be
\[
\tau_R \frac{\partial q}{\partial t} + q = -k \nabla T + \frac{\kappa \tau_c}{c_v} \nabla (\nabla \cdot q). \tag{2.34}
\]
Comparing Eq. (2.34) with Eq. (2.11), we know that \( \tau_B \) in the TPHC model is equivalent to \( \tau_c \) in the EG-BTE, though they are slightly different. Therefore, \( \Lambda_B = c \tau_B = c \tau_c \) still holds. On the other hand, the first term in LHS of Eq. (2.34) is usually negligible. Plugging \( \Lambda_B = c \tau_c \) and \( \Lambda_D = c \tau_D \) into Eq. (2.34) yields
\[
q = -k \nabla T + \frac{\Lambda_D \Lambda_B}{3} \nabla (\nabla \cdot q). \tag{2.35}
\]
We want to emphasize the difference in the nonlocal term between Eq. (2.11) and Eq. (2.35). These two equation are equivalent if one assumption is valid, that is, the heat flux is irrotational, as commonly believed. Thus, the \( \nabla q \) term and \( 2 \nabla (\nabla \cdot q) \) are equivalent so that they can merge into one term \( 3 \nabla (\nabla \cdot q) \). Therefore, this equation is consistent with the TPHC model by Ma [21] as long as the heat flux is irrotational.
It is also due to this derivability that the TPHC model offers almost identical results as by EG-BTE in the scale where system characteristic length $L_c$ is not very small and the nondiffusive effects are not so strong to ruin the validity of the key assumption given by Eq. (2.28). This suggests that they are capturing the nondiffusive heat transfer features in a similar way, and the TPHC model is a simplified case of EG-BTE with the key assumption by Eq. (2.28). As suggested in the objectives section of Chapter 1, the TPHC model mainly focuses on the engineering applications while the EG-BTE mainly focuses on the fundamental study, but they are essentially unified by this derivability. Generally, it is reasonable that there is a tradeoff between the physical details and the computational efficiencies, and this relation between the TPHC model and EG-BTE is a portrayal of the tradeoff: they are not exactly equivalent despite of the demonstrated derivability.

In the end, we specify the range of applicability of these two models and their limitations.

**Targets**  The TPHC model targets the engineering applications, while the EG-BTE focuses more on the fundamental study. For this reason, the TPHC model is mathematically simpler and computationally more efficient than the EG-BTE.

**Range of applicability**  The TPHC model can predict the size-dependence of thermal conductivity in the situations where the nondiffusive effects are relatively weak. In the strong nondiffusive cases where the ballistic phonon transport may be detectable, for example the thermal wave experiments in Chapter 5, EG-BTE has to be applied.

### 2.4 Summary

In this chapter, the Two-Parameter Heat Conduction model is derived based on the definition of nonequilibrium absolute temperature $\beta$. Based on this model, a nondimensional parameter characterizing the applicable range of Fourier’s law is also provided as a criterion for engineering applications. On the other hand, the derivation of Enhanced Gray Boltzmann transport equation is also given starting from the theories of the Boltzmann transport equation for the phonon transport in dielectric materials with certain assumptions. In addition, the derivability of the TPHC model from EG-BTE has also been shown to demonstrate their compatibility.
Chapter 3

Numerical Schemes

In this chapter, numerical schemes for solving the nondiffusive heat transfer models derived in the previous chapter are introduced. The TPHC model can be solved by the regular finite difference method as will be shown, while the EG-BTE has to be solved by specific algorithms due to its particularity. The lattice Boltzmann method is introduced as one of these algorithms. In addition, another algorithm, the discrete ordinates method for phonon transport, is developed in this chapter, with the appropriate boundary condition implementation.

3.1 Numerical scheme for the TPHC model

Due to its mathematical simplicity and direct connection to basic physical quantity, such as temperature, the TPHC model can be solved by the regular finite difference method. Finite difference method is one of the most popular numerical schemes. It is mathematically straightforward and easy to implement. In principle, this method can quickly be specialized for the TPHC model so that a computation package based on the TPHC model for practical engineering projects is available.

Here, without the loss of generality, we only take one-dimensional (1D) situations as an example to explain the scheme. In a two-dimensional (2D) situation, similar treatment can be employed to discretize the equation, and the Alternative Direction Implicit (ADI) method can usually help in converting 2D calculations into quasi-1D. Specifically, to eliminate numerical oscillations and improve accuracy in this study, a staggered grid is implemented.

![Staggered Grid Stencil](image)

Figure 3.1: Stencil for staggered grid.

We start from the 1D TPHC model given by Eqs. (2.3) and (2.9). In the interval $t \rightarrow t + \Delta t$, ...
around the site where \( x = x_j \), the stencil with staggered grid setup is shown in Fig. 3.1. Applying finite difference approximation, we have

\[
\frac{\beta_j^{n+1} - \beta_j^n}{\Delta t} + \frac{q_{j+1/2}^{n+1/2} - q_{j-1/2}^{n+1/2}}{\Delta x} = Q_j^{n+1/2}, \quad (3.1)
\]

\[
q_{j+1/2}^{n+1/2} = -\kappa \frac{\beta_j^{n+1/2} - \beta_j^{n+1/2}}{\Delta x} + \frac{\Lambda D \Lambda B}{3} \frac{d_j^{n+1/2} - 2d_j^{n+1/2} + q_j^{n+1/2}}{\Delta x^2}. \quad (3.2)
\]

Here, \( \Delta t \) and \( \Delta x \) represent for the increment in time and space, respectively. The thermal conductivity \( \kappa \) and ballistic transport length \( \Lambda_B \) are constant. The superscript \( n \) is for time, and the subscript \( j \) is for space. \( n + 1/2 \) in superscript indicates the value of the quantity at certain moment between \( t \) and \( t + \Delta t \). It can generally be obtained by a weighted average

\[
Y^{n+1/2}_j = (1 - \omega_r) Y^n_j + \omega_r Y^{n+1}_j, \quad (3.3)
\]

where \( Y \) can be any quantity (\( \beta \) or \( q \)), \( \omega_r \) is the weight. When \( \omega_r = 0 \), the scheme is explicit. If \( \omega_r = 1 \), the scheme is implicit. When \( \omega_r = \frac{1}{2} \), this scheme is the famous Crank-Nicolson scheme.

When written into matrix form \( AX = b \), where \( X \) is composed of \( \beta \) and \( q \), this TPHC model can be solved by any matrix solver, like the band solver, if appropriate boundary and initial conditions are specified.

The implementation of boundary and initial condition has been specified in Chapter 2. Here we offer an example with a boundary heat flux in 1D situation. Assuming that the heat flux at \( x = 0 \) is given as \( q_w \), a first order discretization of this boundary is to simply replace \( q_j^{n+1/2} \) in Eqs. (3.1) and (3.2), so that we have

\[
\frac{\beta_1^{n+1} - \beta_1^n}{\Delta t} + \frac{Q_1^{n+1/2} - q_w^{n+1/2}}{\Delta x} = Q_1^{n+1/2}, \quad (3.4)
\]

\[
q_{1/2}^{n+1/2} = -\kappa \frac{\beta_2^{n+1/2} - \beta_1^{n+1/2}}{\Delta x} + \frac{\Lambda D \Lambda B}{3} \frac{d_1^{n+1/2} - 2d_1^{n+1/2} + q_w^{n+1/2}}{\Delta x^2}. \quad (3.5)
\]

More details can be found in any computational fluid dynamics (CFD) textbook (i.e., [27]) and thus is not repeated here.

### 3.2 Numerical schemes for the EG-BTE

Due to the existence of equilibrium distribution function \( f^0 \) and the direction-dependence of distribution function, EG-BTE should be solved by some special schemes. We here introduce two schemes designed for solving BTE. The first one is Lattice Boltzmann Method (LBM), while another one is an in-house discrete ordinates method for phonon transport (phonon \( S_N \) method).

#### 3.2.1 Lattice Boltzmann Method

Developed to bridge the gap between microscale and macroscale, the lattice Boltzmann method (LBM) deals with the problem by considering the collective behavior of particles as a whole [28]. In other words, this method is specifically designed to solve mesoscale problems,
which is the purpose of this study. LBM by nature has a number of distinctive advantages over regular methods, such as the clear physical meaning, simple discretized equation structure, natural efficiency for massively parallel architectures and easy implementation of boundary conditions. The applications of LBM are involved in many fields including multi-phase flows with chemical reactions in porous media and combustion [29–31]. In this study, we employ this method for solving EG-BTE.

3.2.1.1 Lattice mesh type

The main idea of LBM is to introduce the discrete direction [32] so that based on the mesh, denoted as lattice, the BTE can be solved. We first introduce the lattice. The lattice type is conventionally marked in the form of $D_mQ_n$, where $m$ is the dimensionality and $n$ is the number of discrete velocities or directions. For example, $D1Q3$ means 1D situation with two discrete velocities (Fig. 3.2(a)). Accordingly, the phonons at one site located at the center of the lattice have three options in a time increment $\Delta t$: propagating to the left, propagating to the right, or staying stagnant at the current site. The discrete velocities in this situation are $v_0 = 0$, $v_1 = 1$, $v_2 = -1$. Similarly, $D2Q9$ means 2D situation with nine discrete velocities (Fig. 3.2(b)). The discrete velocities in this situation are $v_0 = (0, 0)$, $v_1 = (1, 0)$, $v_2 = (-1, 0)$, $v_3 = (0, 1)$, $v_4 = (0, -1)$, $v_5 = (1, 1)$, $v_6 = (-1, -1)$, $v_7 = (-1, 1)$, $v_8 = (1, -1)$. Details can be found in Ref. [33].

![Figure 3.2: Lattice mesh type used in LBM: (a) One dimensional D1Q3 lattice; (b) Two dimensional D2Q9 lattice.](image)

Based on different lattice types, the total energy must be conserved:

$$e(t, x) = \sum_{i=0}^{n} e_i(t, x),$$

(3.6)

where $n$ is the maximal number of discrete directions for a certain lattice type. For instance, $n = 8$ in $D2Q9$. Besides, the equilibrium energy should be appropriately distributed into each discrete directional energy so that the phonon transport can be simulated. Mathematically, this gives

$$e_i^0(t, x) = w_i e(t, x); \quad \text{with} \quad \sum_{i=0}^{n} w_i = 1,$$

(3.7)

where $w_i$ is the weighting factor for $i^{th}$ direction. These weighting factors, along with the speed of sound for the given lattice, can be solved by relating different moments of equilibrium distribution.
functions to the conservation law [33]. Here we only give the results of $D1Q3$ as an example: the sound speed of this lattice arrangement is $1/\sqrt{3}$ of the physical value, and $w_0 = 2/3$, $w_1 = w_2 = 1/6$.

### 3.2.1.2 Lattice Boltzmann kinetic equation

We here use gray BTE given in Eq. (2.25) with $\tau_c = 0$ as an example. EG-BTE can be solved through similar treatment. Gray BTE is given as

$$\frac{\partial e}{\partial t} + \mathbf{v} \cdot \nabla e = -\frac{e - e^0}{\tau_R} + Q.$$  

(3.8)

To solve it, the velocity $\mathbf{v}$ is first discretized based on the lattice type. A projection of distribution function on discrete directions for particles to travel is performed, so that Eq. (3.8) is written as

$$\frac{\partial e_i}{\partial t} + \mathbf{v}_i \cdot \nabla e_i = -\frac{e_i - e^0_i}{\tau_R} + Q_i,$$

(3.9)

where the subscript $i$ stands for different directions, $e_i$ here is called directional phonon energy [22], $e^0_i$ is the directional equilibrium energy, and $\mathbf{v}_i$ is the velocity in the corresponding direction. In 1D situation, the gradient term can be rewritten with discrete velocity. Employing 1D upwind scheme, we have the discretized equation

$$\frac{e_i(t + \Delta t, x) - e_i(t, x)}{\Delta t} + \frac{e_i(t + \Delta t, x + \Delta x) - e_i(t + \Delta t, x)}{\Delta x} = -\frac{e_i - e^0_i}{\tau_R} + Q_i.$$  

(3.10)

Traditionally, there are two steps in LBM named collision step and streaming step, if setting $\Delta t = \Delta x/v$ with .

**Collision step:** $e_i(t + \Delta t, x) = e_i(t, x) - \frac{e_i - e^0_i}{\tau_R}$.

**Streaming step:** $e_i(t + \Delta t, x + v_i \Delta t) = e_i(t + \Delta t, x)$.

For example, in $D1Q2$ lattice, in the streaming step, $e_2(t + \Delta t, x - v_2 \Delta t) = e_2(t + \Delta t, x)$, since $v_2 = -v$. So in the streaming step, $e_i(t + \Delta t, x)$ propagate to the next cite in the direction it is propagating.

The physical implication of these two steps is very clear: phonons are undergoing scattering in collision step and propagating in streaming step. The phonons in LBM are grouped and reside on the discretized sites referred to as lattices. They are allowed to propagate to the neighboring sites in certain discrete directions, which depends on the lattice mesh type as shown in the following section.

### 3.2.1.3 Equilibrium discrete energy density

The equilibrium energy density, equivalently the equilibrium distribution function, is the core part of LBM. Different selections of equilibrium distribution functions, if appropriate, reflect different physical situations. Moreover, the accuracy of an approximated equilibrium distribution function directly determines the accuracy of the simulation results. Viggen [33] showed that the distribution
function can be expanded around equilibrium as
\[ f = f^0 + \epsilon f^1 + \epsilon^2 f^2 + \ldots \] (3.11)
where \( \epsilon \) is a smallness parameter. If the Knudsen number \( Kn = \frac{v}{\tau} \) is defined (\( L \) is the characteristic length of the system), the \( \epsilon \) indicate that \( f^1/f^0 = O(Kn) \), \( f^2/f^0 = O(Kn^2) \). Again, by relating each order moment of the equilibrium distribution function to the conservation law, \( f^0 \) can be solved. Then, the discrete equilibrium distribution function can be calculated using the weighting factor
\[ f_i^0 = w_i f^0. \] (3.12)

However, it should be noted that in the cases where the distribution function is very close to equilibrium (i.e., \( Kn \) is very small), \( f \approx f^0 \). Hence, \( e_i^0 = w_i e^0 = w_i \sum_{j=0}^n e_j \). For the cases where the distribution function has significant deviation from equilibrium, this low order formula does not hold anymore, and a high order formula has to be applied. Unfortunately, distinctive features of phonon transport from regular mass transport in fluid mechanics hinder the process to acquire a high order formula from traditional methods. For example, momentum is not conserved during the U process scattering, as introduced in Chapter 1, posing issues against the conventional way to apply conservation laws to calculate the equilibrium function.

At present, the equilibrium function is still approximated as \( f^0 \approx f \). This formula becomes questionable when \( Kn \) is not small enough. High order formula for equilibrium function remains a topic for our future study.

### 3.2.1.4 Implementation of boundary conditions

The implementation of boundary conditions is straightforward in LBM due to its clear physical picture. Mathematically, there are three types of boundary conditions: the Dirichlet (or first-type) boundary condition, the Neumann (or second-type) boundary condition, and the Robin (or third-type) boundary condition. In heat transfer, the Dirichlet boundary condition indicates a known boundary temperature; the Neumann boundary condition indicates a known boundary heat flux; and the Robin boundary condition indicates a mixture, such as convective heat transfer or thermal interface. Based on the relations listed in the previous section (Eq. (2.26) and Eq. (2.27)), these boundary conditions can be adapted to LBM.

**Dirichlet boundary condition** The boundary temperature is specified here. According to Eq. (2.26) and the approximation \( e \approx e^0 \) derived from \( f \approx f^0 \), there exists
\[ T(t) = \frac{4\pi}{c_v} e^0 \approx \frac{4\pi}{c_v} \sum_{j=0}^n e_j. \] (3.13)

Thus, in \( D1Q3 \) lattice at \( x = 0 \) for example, the boundary condition for \( e_1(t, x = 0) \) is
\[ e_1(t, x = 0) = \frac{c_v}{4\pi} T(t) - e_0(t, x = 0) - e_2(t, x = 0). \]

**Neumann boundary condition** The boundary heat flux is specified here. According to Eq. (2.27),
there exists
\[ q(t) = c_v \sum_{i=1}^{n} v_i e_i. \] (3.14)

Thus, in the $D1Q3$ lattice at $x = 0$ for example, the boundary condition for $e_1 (t, x = 0)$ is
\[ e_1 (t, x = 0) = \frac{q(t)}{c_v} - (-ve_2). \]

**Robin boundary condition** The relation between boundary temperature and heat flux is specified here, $AT \cdot B \cdot q = 0$ for example. As a combination of previous two kinds of boundary conditions, the boundary condition of the third kind can be expressed as
\[ AT (t) + B \cdot q(t) = A \frac{4 \pi}{c_v} \sum_{i=0}^{n} e_i + B \cdot c_v \sum_{i=1}^{n} v_i e_i. \] (3.15)

Thus, in $D1Q3$ lattice at $x = 0$ for example, the boundary condition for $e_1 (t, x = 0)$ is
\[ \left( A \frac{4 \pi}{c_v} + B c_v \right) e_1 (t, x = 0) = -A \frac{4 \pi}{c_v} e_0 (t, x = 0) - \left( A \frac{4 \pi}{c_v} - B c_v \right) e_2 (t, x = 0). \] (3.16)

Finally, the implementation of the initial condition is the same as in the regular finite difference method. In fact, it is the finite difference method that is applied after the discretization of velocity on a lattice for solving the discrete equations.

After the implementation of boundary and initial conditions, we introduce an implicit LBM for solving the EG-BTE in the next subsection.

### 3.2.1.5 The implicit lattice Boltzmann method

As mentioned before, the conventional explicit LBM divides particle transport into two steps: collision step and streaming step. However, this explicit LBM is unconditionally unstable for EG-BTE according to Wang et al. [22]. As a consequence, Wang et al. proposed an implicit lattice Boltzmann method (ILBM) that is unconditionally stable for EG-BTE [22]. Assuming the material is isotropic, under the framework of LBM, Eq. (2.25) can be discretized as
\[
\frac{e_i(t + \Delta t, \mathbf{r}) - e_i(t, \mathbf{r})}{\Delta t} + v_i \frac{e_i(t + \Delta t, \mathbf{r} + \Delta \mathbf{r}_i) - e_i(t + \Delta t, \mathbf{r})}{\Delta \mathbf{r}_i} = \frac{-e_i(t + \Delta t, \mathbf{r}) - e_i(t, \mathbf{r})}{\tau_R} - \frac{e_i(t + \Delta t, \mathbf{r}) - e_i(t, \mathbf{r})}{\tau_c v_i} \frac{e_i(t + \Delta t, \mathbf{r} + \Delta \mathbf{r}_i) - e_i(t + \Delta t, \mathbf{r}) - e_i(t, \mathbf{r} + \Delta \mathbf{r}_i) + e_i(t, \mathbf{r})}{\Delta t \Delta \mathbf{r}_i}. \] (3.17)

Eq. (3.17) shows the general form of ILBM. A detailed example on a $D2Q4$ lattice is given in Ref. [9].

Different from the explicit LBM, the ILBM is free from the constraint $\Delta \mathbf{r}_i = v_i \Delta t$, and has been proven to be unconditionally stable. This allows the free choice of $\Delta t$ and $\Delta \mathbf{r}_i$, facilitating fast
computation on grid-independent results. However, it should be pointed out that, a matrix solver or an iterative method is required, which impedes parallelization, and thus may actually undermine the computation performance. In fact, the main purpose for developing this ILBM is to avoid diverging results when using explicit method to solve the EG-BTE.

### 3.2.1.6 Summary for LBM

LBM has been described in this section. Based on discrete velocities according to selected lattice type, directional phonon energy can be solved on each lattice site. Next, the equilibrium phonon energy can be obtained by summing up all the directional components. The solved equilibrium phonon energy can be related to the temperature, and the simulation proceeds to the next time step until a desired moment is reached. A procedure for simulation flowchart is given in Fig. 3.3.

![Simulation procedure flowchart for LBM.](image-url)

Figure 3.3: Simulation procedure flowchart for LBM.
3.2.2 Discrete ordinates method for phonon transport

Besides the LBM, a most straightforward strategy to solve a directional partial differential equation like Eq. (3.8) is to discretize the directions first, which is also one main idea of LBM. The same idea has already initiated the development of numerical algorithm for solving the radiative transfer equation (RTE). For example, the spherical harmonics method and the discrete ordinate method are two of the most popular RTE solvers. They both are tools to transform the equation of transfer into a set of simultaneous partial differential equations [34]. Although the BTE differs from the RTE, the direction-dependence property of the BTE can also be solved with adapted schemes. In the previous work [35], the spherical harmonics method was implemented for the derivation of the enhanced Fourier law. In this study, a simplified adaptation of discrete ordinates method for phonon transport (phonon $S_N$ method) is presented, and its applications will be shown in the following chapters.

3.2.2.1 Discrete ordinates equations

In the LBM, the approximation for the equilibrium distribution function does not hold when Kn is not small enough. However, as mentioned in Chapter 2, in a gray medium the distribution function $f$ can be related to the equilibrium distribution function $f^0$

$$f^0 = \frac{1}{4\pi} \int_{4\pi} f d\Omega.$$  \hspace{1cm} (3.18)

In 1D situation, assuming the distribution function to be azimuthally symmetric, we have

$$f^0 = \frac{1}{2} \int_{0}^{\pi} f(\theta) \sin \theta d\theta.$$  \hspace{1cm} (3.19)

If the integral in Eq. (3.19) is solved numerically for a set of $N$ discrete directions, we have

$$f^0 = \frac{1}{2} \int_{0}^{\pi} f(\theta) \sin \theta d\theta \approx \frac{1}{2} \sum_{i=1}^{N} f(\theta_i) \sin \theta_i \Delta \theta = \sum_{i=1}^{N} w_i f_i,$$  \hspace{1cm} (3.20)

or

$$e^0 = \frac{1}{2} \int_{0}^{\pi} e(\theta) \sin \theta d\theta \approx \frac{1}{2} \sum_{i=1}^{N} e(\theta_i) \sin \theta_i \Delta \theta = \sum_{i=1}^{N} w_i e_i,$$  \hspace{1cm} (3.21)

where the $w_i$ are the weights for each discrete direction. Compared with Eq. (3.7), Eq. (3.21) is a more general form with a better directional approximation since the numerical quadrature can be solved with arbitrary accuracy in theory. This relation serves as a replacement for the calculation of the equilibrium distribution function, or equilibrium phonon energy at each grid site, making it free from the restriction that Kn has to be adequately small. Accordingly, the heat flux given by Eq.
(2.27) in 1D can also be approximated as

$$q_x = \int_0^{2\pi} \mathbf{v} \cdot \mathbf{s}_i d\Omega = \int_0^{2\pi} d\phi \int_0^{\pi} \mathbf{v} \cos \theta \sin \theta d\theta = 2\pi \int_0^{\pi} \mathbf{v} \cos \theta \sin \theta d\theta$$

$$= 2\pi \int_0^{\pi} \mathbf{v} \cos \theta \sin \theta d\theta = 2\pi \sum_{i=1}^n e_i \sin \theta_i \cos \theta_i \Delta \theta. \quad (3.22)$$

This opens up the possibility to apply this method for highly nonequilibrium cases where the thermal transport is ballistic, which will be shown in Chapter 5.

Thus, the directional BTE can be approximated by $N$ equations in all $N$ discrete directions. Without loss of generality, we still take the 1D gray BTE in energy form as an example:

$$\frac{\partial e_i}{\partial t} + v \cos \theta_i \frac{\partial e_i}{\partial x} = -\frac{e_i - e^0}{\tau_R} + Q. \quad (3.23)$$

Integrating this equation by segments from $\theta_i - \Delta \theta/2$ to $\theta_i + \Delta \theta/2$

$$\int_{\theta_i - \Delta \theta/2}^{\theta_i + \Delta \theta/2} \left( \frac{\partial e_i}{\partial t} + v \cos \theta_i \frac{\partial e_i}{\partial x} \right) d\theta = \int_{\theta_i - \Delta \theta/2}^{\theta_i + \Delta \theta/2} \left( -\frac{e_i - e^0}{\tau_R} + Q \right) d\theta. \quad (3.24)$$

Since $e^0$ is isotropic and external source $Q$ is assumed to be isotropic, they can be taken out of the integral. Approximating this integral yields

$$\left( \frac{\partial e_i}{\partial t} + v \cos \theta_i \frac{\partial e_i}{\partial x} \right) \Delta \theta = \left( -\frac{e_i - e^0}{\tau_R} + Q \right) \Delta \theta, \quad (3.25)$$

which can be rewritten by canceling out $\Delta \theta$ as

$$\frac{\partial e_i}{\partial t} + v \cos \theta_i \frac{\partial e_i}{\partial x} = -\frac{e_i - e^0}{\tau_R} + Q. \quad (3.26)$$

If the total number of discrete directions $N$ is 2, Eq. (3.26) coincides with the lattice Boltzmann kinetic equation on the $D1Q2$ lattice arrangement.

Adding up all $N$ equations in each discrete direction, together with Eq. (3.19), we obtain $N + 1$ equations for $N + 1$ unknown variables: $e_1, \ldots, e_N, e^0$. Thus, the problem is mathematically closed.

The next step is to solve these $N + 1$ partial differential equations, which involves an upwind finite difference method.

### 3.2.2.2 Upwind finite difference scheme

The EG-BTE by nature shows wave behavior, indicating that it is necessary to employ an upwind scheme to obtain a converged solution. We first give the first order upwind scheme here.

Discretizing the $i^{th}$ equation in time and space by finite differences yields

$$\frac{e_{i,j}^{n+1} - e_{i,j}^n}{\Delta t} + v \cos \theta_i \frac{e_{i,j}^{n+1} - e_{i,j-1}^{n+1}}{\Delta x} = -\frac{e_j^n - e_{i,j}^n}{\tau_R} + Q_j^{n+1}, \quad (3.27)$$

$$\frac{e_{i,j}^{n+1} - e_{i,j}^n}{\Delta t} + v \cos \theta_i \frac{e_{i,j}^{n+1} - e_{i,j-1}^{n+1}}{\Delta x} = -\frac{e_j^n - e_{i,j}^n}{\tau_R} + Q_j^{n+1}, \quad (3.27)$$
where the subscript \( i \) stands for \( i^{th} \) discrete direction, \( j \) represents \( j^{th} \) grid point, and the superscript \( n \) means \( n^{th} \) time step. Equation (3.27) is valid for all the discrete directions with \( v_i \cdot \hat{x} > 0 \), where \( \hat{x} \) is a unit vector pointing into the positive \( x \) direction. For the remaining discrete directions, we have

\[
\frac{e_{i,j}^{n+1} - e_{i,j}^n}{\Delta t} + v \cos \theta_i \frac{e_{i,j+1}^{n+1} - e_{i,j}^{n+1}}{\Delta x} = \frac{e_{j}^{0,n+1} - e_{i,j}^{n+1}}{\tau_R} + Q_{j}^{n+1}.
\]

(3.28)

Eq. (3.27) and Eq. (3.28) can be solved by iteration.

Similarly, the second order upwind scheme, which is the one we use in later chapters, is given as

\[
\frac{e_{i,j}^{n+1} - e_{i,j}^n}{\Delta t} + v \cos \theta_i \frac{\frac{3}{2}e_{i,j+1}^{n+1} - 2e_{i,j}^{n+1} + \frac{1}{2}e_{i,j-1}^{n+1}}{\Delta x} = \frac{e_{j}^{0,n+1} - e_{i,j}^{n+1}}{\tau_R} + Q_{j}^{n+1},
\]

(3.29)

for positive discrete directions, and

\[
\frac{e_{i,j}^{n+1} - e_{i,j}^n}{\Delta t} + v \cos \theta_i \frac{-\frac{3}{2}e_{i,j+1}^{n+1} + 2e_{i,j}^{n+1} - \frac{1}{2}e_{i,j+2}^{n+1}}{\Delta x} = \frac{e_{j}^{0,n+1} - e_{i,j}^{n+1}}{\tau_R} + Q_{j}^{n+1}.
\]

(3.30)

for negative discrete directions.

A higher order upwind scheme for solving these equation can be similarly obtained by replacing the discretization of the derivative with higher order finite differences.

However, it should be noted that the lower order upwind scheme (like first order) shows strong numerical dissipation, while the higher order upwind scheme (like third order) on the contrary shows strong dispersion effects. Neither dissipation nor dispersion is desired and has to be eliminated by applying more advanced numerical algorithms. The purpose of this study is to discover the physics in phonon transport, so once proved to have acceptable accuracy, the lower order scheme is preferred due to lower computation expenses. In general, the second order upwind scheme satisfies our requirement well in this study.

Having prepared the discretized governing equations, we move on to the implementation of boundary conditions. The implementation of initial conditions, on the other hand, is standard and not repeated here.

3.2.2.3 Implementation of boundary conditions

Boundary conditions are sometimes important factor for certain engineering problem. For instance, boundary scattering in thermal transport processes inside thin films can severely influence the ETC, as has been observed in 1D TTG on silicon thin films. Unfortunately, the details of phonon boundary scattering are still not well understood despite numerous studies. Here, based on the most simplified and widely used assumptions, we introduce the boundary conditions for the phonon \( S_N \) method.

When phonons travel inside a bulk material, known as bulk phonons, collide with the boundary, there are three available options: transmission, reflection or becoming surface phonons. Surface phonons are the particular modes associated with surfaces due to the termination of crystal structure. They propagate along the surface and their amplitudes decay towards the bulk. Among them, the Rayleigh wave is a special mode which survives up to the continuum limit, and will be mentioned in
the following chapters. The transition from bulk phonons to surface phonons may have significant impact to our boundary condition discussed here, but is not considered here due to lack of understanding. In this study, this conversion is assumed negligible, as commonly believed. Thus, only two options for phonons are left: transmission or reflection. This is very similar to photon transport in radiation, so the concept of diffuse or specular wall can be applied here.

**Diffuse** A diffuse boundary results in random reflection or transmission of phonons. A diffuse wall suggests loss of memory: the traveling direction of the phonon reflected by the wall is independent of its incident angle, or the phonon “forgets” its history in other words. Moreover, the phonon forgets whether it was reflected from one side of the interface or transmitted from the other side of the interface, if transmission is possible. Such a definition introduces an isotropic behavior so that the reflected phonon density is equally distributed in all discrete directions.

**Specular** A specular boundary results in reflection or transmission that satisfies the laws of optics. For example, the tangent component velocity of the phonon reflected by the wall remains unchanged, while the normal component is reversed.

Practical situations should fall between these two extreme cases, and are influenced by many factors such as surface roughness and temperature. Here, we only consider boundary conditions for these two cases.

**Dirichlet boundary condition** The wall temperature is specified. The specular assumption is usually not applicable since it requires the input of information (i.e., incident angle) from the other side, which is unknown unless given. Once the energy distribution over the solid angle is specified, $e_i$ on $i^{th}$ discrete direction can be obtained by integrating the distribution over the corresponding solid angle. For the diffuse case, phonons emitted by the wall or transmitted from the other side are isotropic. Therefore, according to Eq. (2.26) and Eq. (3.21), given the specified wall temperature $T_w$, we have

$$T_w = \frac{4\pi}{c_v} \left( \int_0^{\pi/2} \frac{1}{2} e_i|_{x=0} \sin \theta d\theta + \int_{\pi/2}^{\pi} \frac{1}{2} e_i|_{x=0} \sin \theta d\theta \right)$$

$$= \frac{4\pi}{c_v} \left( \frac{1}{2} \sum_{i=1}^{N/2} e_i|_{x=0} \sin \theta_i \Delta \theta + \sum_{i=N/2+1}^{N} \frac{1}{2} e_i|_{x=0} \sin \theta_i \Delta \theta \right). \quad (3.31)$$

Since EG-BTE is a first order PDE in space, only one boundary condition is needed for each discrete equation. Among these equations, only those with positive velocity need boundary conditions, if we take the boundary at $x = 0$ as an example. Therefore, the boundary condition for these discrete equations is expressed as

$$e_i|_{x=0} = \frac{e_i T_w}{2\pi} - \frac{\sum_{j=N/2+1}^{N} e_j|_{x=0} \sin \theta_j \Delta \theta}{\sum_{j=1}^{N/2} \frac{1}{2} \sin \theta_j \Delta \theta} \quad \text{for} \ i = 1, \ldots, N/2 \quad (3.32)$$

where $N$ can always be chosen to be an even number. Eq. (3.32) is also applicable for a known time-dependent wall temperature, as in the thermal wave problem in Chapter 5.

**Neumann boundary condition** The heat flux from the wall is specified here. We first provide the corresponding form based on specular assumption at $x = L$ for example. Using the specular
assumption, however, input information such as the incident angle is still necessary to maintain the traveling direction of the transmitted phonons. Again, such information is usually unavailable. For practical use, an insulated wall that covers a wide range for engineering problems is considered here:

\[
e_i|_{x=L} = (1 - \alpha) e_{N-i+1}|_{x=L} \text{ for } i = \frac{N}{2} + 1, \ldots, N,
\]

where \(\alpha\) is the transmissivity. If the wall is insulated, \(\alpha = 0\).

The other limit is a diffuse wall. The directional phonon energy is isotropic so that according to Eq. (3.22), there exists

\[
q_w = 2\pi v \int_{0}^{\pi/2} e|_{x=L} \sin \theta \cos \theta d\theta + 2\pi v \int_{\pi/2}^{\pi} e|_{x=L} \sin \theta \cos \theta d\theta
\]

\[
= 2\pi v \sum_{i=1}^{N/2} e|_{x=L} \sin \theta_i \cos \theta_i \Delta \theta + 2\pi v \sum_{i=N/2+1}^{N} e|_{x=L} \sin \theta_i \cos \theta_i \Delta \theta
\]

(3.34)

Since \(e|_{x=L}\) under diffuse assumption is independent of solid angle, the solution for the part of \(e|_{x=L}\) is expressed as

\[
e|_{x=L} = \frac{q_w - 2\pi v \sum_{j=1}^{N/2} e_{j}|_{x=L} \sin \theta_j \cos \theta_j \Delta \theta}{2\pi v \sum_{j=N/2+1}^{N} \sin \theta_j \cos \theta_j \Delta \theta}, \text{ for } i = \frac{N}{2} + 1, \ldots, N,
\]

(3.35)

where \(q_w\) is the specified heat flux from the wall.

**Robin boundary condition** The relation between boundary temperature and heat flux is specified here. We take the convection form \(q_w = h_c (T - T_{\infty})\) at \(x = L\) as an example, where \(h_c\) is the convective heat transfer coefficient, or the thermal conductance of the interface. As a combination to previous two kinds of boundary conditions, the third kind boundary condition under diffuse assumption can be expressed as

\[
\int_{0}^{\pi/2} e|_{x=L} \sin \theta \cos \theta d\theta + \int_{\pi/2}^{\pi} e|_{x=L} \sin \theta \cos \theta d\theta
\]

\[
= H_c \left(\int_{0}^{\pi/2} \frac{1}{2} e|_{x=L} \sin \theta d\theta + \int_{\pi/2}^{\pi} \frac{1}{2} e|_{x=L} \sin \theta d\theta\right) - H_c T_{\infty},
\]

(3.36)

where \(H_c = h_c/2\pi v\). After discretization, we have

\[
\sum_{i=1}^{N/2} e_{i}|_{x=L} \sin \theta_i \cos \theta_i \Delta \theta + \sum_{i=N/2+1}^{N} e_{i}|_{x=L} \sin \theta_i \cos \theta_i \Delta \theta
\]

\[
= H_c \left(\sum_{i=1}^{N/2} \frac{1}{2} e_{i}|_{x=L} \sin \theta_i \Delta \theta + \sum_{i=N/2+1}^{N} \frac{1}{2} e_{i}|_{x=L} \sin \theta_i \Delta \theta\right) - H_c T_{\infty}.
\]

(3.37)
Thus, $e_i|_{x=L}$ can be solved as

$$e_i|_{x=L} = \sum_{j=1}^{N/2} e_j|_{x=L} \left( \frac{H_i}{2} - \cos \theta_j \right) \sin \theta_j \Delta \theta$$

for $i = \frac{N}{2} + 1, \ldots, N$. (3.38)

Eq. (3.32), (3.35) and (3.38) are the three kinds of boundary conditions for phonon $S_N$ method under the diffuse assumption.

### 3.2.2.4 Summary for the phonon $S_N$ method

The discrete ordinates method for phonon transport has been described in this section. With variables discretized based on the $N$ selected discrete directions, a connection between the directional phonon energy and the equilibrium phonon energy is established. This relation compared to the approximation implemented in LBM is more accurate and has the potential to reach arbitrary order of accuracy. Accordingly, the boundary conditions for phonon $S_N$ method have been developed, preparing the phonon $S_N$ method for numerical simulation. Iterative algorithm should be employed for solving EG-BTE through phonon $S_N$ method. A procedure of simulation flowchart is given in Fig. 3.4

### 3.3 Summary

In this chapter, three numerical schemes that will be applied to solve TPHC model and EG-BTE derived in Chapter 2 are introduced. Finite difference method with staggered grid is introduced as the first one for numerically solving TPHC model, followed by the other two designed for EG-BTE. One of these two is the lattice Boltzmann method (LBM), the other is the discrete ordinates method for phonon transport (phonon $S_N$ method). The simulation algorithm as well as the boundary conditions setup has been described in details. The procedure flowcharts for both methods are also given in the corresponding summary parts.
Set initial phonon energy density at each site

Evolve to the next time step

Calculate energy density in each discrete direction at each site

Calculate the equilibrium phonon energy density at each site

Converged?

Update new phonon density at each site

Reach desired time?

Start

Yes

End

No

Yes

No

Figure 3.4: Simulation procedure flowchart for discrete ordinates method.
Chapter 4

Application of the Two-Parameter Heat Conduction Model

In this chapter, the Two-Parameter Heat Conduction (TPHC) model is applied on multiple nondiffusive heat transfer experiments, namely 1D/2D transient thermal grating (TTG) experiment, and metallic grating experiment measured by diffracted X-ray (MG-X-ray experiment). The size-dependent nondiffusive heat transfer observed in the experiments can be captured by the TPHC model. In addition, the criterion based on the nondimensional parameter \( \zeta \) defined in Chapter 2 is also illustrated, showing the potential for engineering applications.

4.1 Applications of the TPHC model on the 1D TTG experiments

Although there exist many experimental observations of nondiffusive heat transfer, a quantitative measurement of nondiffusive heat transfer is still not an easy task. Among all experimental techniques, transient thermal grating (TTG) is one of the quantitative measurements that can capture the size-dependence of nondiffusive heat transfer. This technique, according to Johnson et al. [36], has three important advantages: (1) there exists no thermal interfaces; (2) the thermal transport is 1D; (3) the heat transfer characteristic length is controllable. These features make the TTG experiment an outstanding quantitative observation of nondiffusive heat transfer and a validation for the nondiffusive heat transfer model.

4.1.1 Physical situation of 1D TTG experiments

In TTG experiments conducted by Johnson et al. [37], the experimental setup for the experiment involves a dielectric sample that can either be a thin film to approximate 1D, or a substrate to explore 2D heat transfer. The sample is heated by two concurrent interfering pulsed-lasers, leading to a spatially periodic micron-sized thermal grating (periodic spaced hot and cold regimes). The spatial grating size of the thermal grating \( P \) can be determined by the laser wavelength \( \lambda_e \) and the crossing angle \( \theta_e \) between the two interfering lasers:

\[
P = \frac{\lambda_e}{2 \sin (\theta_e/2)}.
\]
In 1D TTG, the sample is very thin, so that the probe laser for taking the measurements is recorded on the other side of the sample due to the transmission; while in 2D TTG, the signal is reflected and recorded from the same side as the heating. The measured signal by the probe laser is the time evolution of the temperature field, noted as the thermal decay.

Physically, the heating process is equivalent to an volumetric heat generation (source term) in the sample. During the laser heating, electrons are excited first. The electrons will pass thermal energy to phonons through an electron-phonon interaction process. This heating process happens so fast (within the 60 ps pulse duration) compared to the recording time of the experiment (hundreds of ns) that can be approximated by a Dirac delta function in time. Spatially, this source term is a sinusoidal distribution in the in-plane (parallel to the sample surface) direction, which will be later referred to as $x$ direction. In the cross-plane direction (normal to the sample surface, referred to as $y$ direction), the temperature change decays exponentially. Specifically, for 1D TTG with a thin film sample, no significant variations exist in the cross-plane direction, which is the reason for thin film TTG to be treated as 1D. After the heating process, the rise of temperature due to pump laser heating is detected by a diffracted signal from the probe laser. This probe laser, however, is also characterized by the its penetration depth into the sample, just as the pump heat laser. In the 1D TTG experiment, it is the temperature field that can directly be measured by the probe laser since the penetration depth in this situation is much larger than the film thickness. Figure 4.1 shows a schematic view of the physical situation in the 1D TTG experiment. The experimental results are shown in Fig. 4.1(b). In this figure, the measured effective thermal conductivities (ETCs) normalized by bulk thermal conductivity of silicon at different grating period $P$ are plotted. Two silicon thin film samples with similar thickness (membrane 1: 392 nm, membrane 2: 390 nm) are measured in the experiment. The results indicate that the ETC increases almost linearly in the regime where $P < 5 \mu m$ and gradually converges to a constant in the regime where $P > 15 \mu m$. These results serve as direct evidence for nondiffusive heat transfer.

4.1.2 Analytical solution by the TPHC model

According to the physical description of the 1D TTG experiment, the mathematical description of this experiment can be determined. Assuming that the spatial period size in $x$ direction is $P$,
we can specify the source term in Eq. (2.3) in the 1D TTG experiment as

\[ Q(t, x) = Q_0 \exp \left[ i \left( \frac{2\pi}{P} \right) x \right] \delta(t), \]  

(4.2)

where \( Q_0 \) is the amplitude of this term, which influences the exact value of the maximal temperature rising, but does not affect the major results such as thermal decay rate and ETC. For mathematical simplicity, we take \( Q_0 = 1 \), because in the experimental results given by Johnson et al., the signal is also normalized to its maximum.

Given the source, the governing equations for 1D TTG experiment based on the TPHC model can be specialized from Eqs. (2.3) and (2.9) to

\[ c_v \frac{\partial \beta}{\partial t} + \frac{\partial q}{\partial x} = \exp \left[ i \left( \frac{2\pi}{P} \right) x \right] \delta(t), \]  

(4.3)

\[ q = -\kappa \frac{\partial \beta}{\partial x} + \frac{\Lambda_D \Lambda_B}{3} \frac{\partial^2 q}{\partial x^2}. \]  

(4.4)

The boundary condition in this case is periodic due to the heating pattern.

Equations (4.3) and (4.4) can be solved on frequency domain after Fourier transform and inverted back to the time and space domain. But an alternative solution is to assume a trial solution that takes form \( \beta = \beta_0 \exp (i\lambda x - \gamma t) \) and \( q = q_0 \exp (i\lambda x - \gamma t) \), where \( \lambda = 2\pi/P \) is the wavenumber of the spatial grating and \( \gamma \) is the thermal decay rate in 1D TTG. Substituting these expressions into Eqs. (4.3) and (4.4), the thermal decay rate can be solved as

\[ \gamma = \frac{\lambda^2 \kappa/c_v}{1 + \frac{\Lambda_D \Lambda_B}{3} \lambda^2} = \frac{\lambda^2 \kappa/c_v}{1 + \zeta}, \]  

(4.5)

where \( \zeta \) is the nondimensional parameter characterizing nondiffusive heat transfer defined in Eq. (2.12), with the system characteristic length \( L_c = 1/\lambda = P/2\pi \). Comparing Eq. (4.5) to the solution by Fourier’s law, which can be obtained by setting \( \Lambda_B = 0 \), the ETC in 1D TTG can be expressed as

\[ \kappa_{\text{eff}} = \gamma c_v/\lambda^2 = \frac{\kappa_{\text{diff}}}{1 + \frac{\Lambda_D \Lambda_B}{3}} = \frac{\kappa_{\text{diff}}}{1 + \zeta}, \]  

(4.6)

where \( \kappa_{\text{diff}} \) is the diffusive thermal conductivity differing from \( \kappa_{\text{bulk}} \). This is because the thermal conductivity of a thin film is different from the bulk with the same material even when diffusive assumption is valid.

Eq. (4.6) relates ETC to the nondiffusive effects characterized by \( \zeta \), and consequently introduces the size-dependence of ETC: as the nondiffusive effects become stronger in smaller length scales, the ETC decreases to a lower value. Eq. (4.6) also offers a quantitative description on the limit of applicable scope of Fourier’s law. For example, if 1% change in thermal conductivity is considered as large, a critical value of \( \zeta_c = 0.01 \) can be selected. Similarly, if 5% change is considered as large, \( \zeta_c = 0.052 \). This criterion should be related to the accuracy requirement of the engineering applications. In this thesis, we keep \( \zeta_c = 0.01 \) as an example.

Unfortunately, the ETC cannot always be quantitatively related to \( \zeta \) as in this 1D TTG case. We will show later that in 2D TTG and MG-X-ray experiments, there exists no explicit formula of ETC due to multidimensional heat transfer. An anisotropic ETC is able to fit the measured signal as
well as an isotropic ETC, questioning the mathematical uniqueness of the fitted ETC and thus the framework of Fourier’s law in this scale.

4.1.3 Comparison with experiments

Johnson et al. [36] performed 1D TTG experiments on silicon thin films, as noted at the beginning of this chapter. To match the experiments, the material’s thermophysical properties are given as: volumetric specific heat $c_v = 1.66 \times 10^6 \text{J/m}^3\text{K}$, speed of sound (Debye velocity) $v = 6400 \text{m/s}$ [38]. The diffusive thermal conductivity for the thin film was measured as $\kappa = 92 \text{W/mK}$ with large grating period $P$, which is different from the bulk value of $\kappa_{\text{bulk}} = 148 \text{W/mK}$.

We first calculate the value of key parameters in the TPHC model. From kinetic theory, $\kappa = c_v \frac{\Lambda_D}{v^2}$ so that the diffusive MFP $\Lambda_D$ is calculated as $\Lambda_D = 3k/c_vv = 26 \text{nm}$. On the other hand, the ballistic transport length $\Lambda_B$ has been fitted by Ma [21] as $\Lambda_B = 7.1 \mu\text{m}$. Plugging in these numbers, Eqs. (4.6) and (4.5) provide the ETC and thermal decay rate for 1D TTG experiments, respectively.

Figure (4.2) displays the comparison between theoretical results by the TPHC model and experimental observations. In Fig. 4.2(a) the thermal decay rate $\gamma$ calculated by the TPHC model is in excellent agreement with the experimental measurements, while Fourier’s law departs from the measurements when $\lambda^2$ increases. On the other hand, the ETC calculated by the TPHC model using Eq. (4.6) is compared with experiments as well as results by some other nondiffusive models in Fig. 4.2(b). In this figure, the considerably large deviation by gray BTE and the nice match by the TPHC model distinguish each other, indicating a better accuracy of the TPHC model in this situation compared to gray BTE. On the other hand, the theoretical results that consider the “correction factor” report by Johnson et al. are plotted as the dash-dot line. Both this theory and the TPHC model explain the experiments appropriately in intermediate and diffusive regime where $P$ is approximately larger than 4 $\mu\text{m}$, while the TPHC model fits the experiments a little better in the nondiffusive regime.

![Figure 4.2](image-url)

**Figure 4.2:** (a). The thermal decay rate and (b). size-dependent effective thermal conductivity of silicon thin films measured from 1D TTG varies with thermal grating periods.
In addition to the extracted ETC and thermal decay rate, a direct comparison of the signal, given by Ma [21], between experiment and our analytical solutions are also plotted in Fig. 4.3. As expected, they agree well with each other. In this figure, however, the initial rising of the signal is fitted to the experiment. In detail, we identify the peak of the signal and start our calculation at the corresponding moment, and fit the rising part of the curve by interpolation. Combining these two parts gives a complete matched signal. This is because the absorption during the laser heating is not in the applicable scope of our model. In the initial heating stage, the heat transfer behaves strongly nondiffusively or even ballistically as indicated by Collins et al. [39]. Thus, it is not appropriate to construct the characterization of heat transfer under the framework of thermal conductivity. Although the TPHC model is not applicable at this stage, EG-BTE still can predict some features of thermal transport here, as will be shown in Chapter 5.

4.2 Applications of the TPHC model on 2D TTG experiments

In addition to 1D TTG experiment on silicon thin film, Johnson et al. also performed reflection TTG measurements of thermal transport in a bulk GaAs sample [37, 40], where there is 2D heat transfer. We first derive an analytical solution based on the TPHC model in this section, and compare this solution to the both the existing experimental results and the spectral BTE solutions to validate it. In addition, the nondimensional parameter $\zeta$ is extended to multidimensional situations (i.e., 2D for this case) to characterize nondiffusive heat transfer in these situations.

As mentioned in 1D TTG experimental setup part, the experimental setup for 2D TTG is very similar to that of 1D TTG except for two major differences. The first one is the sample measured in the experiment is bulk GaAs instead of silicon thin films. The second difference is that the diffracted signal is taken from the heating side as reflection. Therefore, the schematic of the 2D TTG experiments is not given here.
4.2.1 Physical situation of 2D TTG experiments

Accordingly, there are two major differences in the physical situation of 2D TTG from 1D TTG experiments. The first one that corresponds to the sample is that, the heat transfer here is 2D instead of 1D due to the finite penetration depth of the heating laser into the material. The energy of the laser pump entering the material is absorbed on its path by the material through their interactions, such as photon-phonon and photon-electron scatterings. The second difference is the interpretation of the signal: it is no longer simply the temperature profile in the sample but a weighted average over the temperature profile in the cross-plane direction at the corresponding location. This is also because of the finite penetration depth of the probe laser.

Mathematically, the spatially sinusoidal source term implemented in 1D TTG still holds in the in-plane direction, while in the cross-plane direction the source term decays exponentially into the depth according to Beer-Lambert law. Similar to 1D TTG, we consider that the source term is approximately instantaneous.

4.2.2 Analytical solution by the TPHC model

According to the physical description given above, the source term can be written as

\[ Q(t, x, y) = Q_0 \exp (i \lambda x - \beta_1 |y|) \delta(t), \]  

(4.7)

where \( \beta_1 \) corresponds to the absorption coefficient of the pump beam in the material, and \( \lambda = 2\pi/P \) still represents the wavenumber of the thermal grating pattern. We still set \( Q_0 = 1 \).

The boundary conditions (BC) for this situation in the in-plane direction are still periodic as in 1D TTG. On the material surface of the heating side, the boundary is considered as insulated, because the time scale is so short that energy loss is usually considered as negligible; while on the other side, the domain is considered as infinity due to the large thickness of the bulk material.

We first take the divergence on both sides of Eq. (2.11)

\[ \nabla \cdot \mathbf{q} = -\nabla \cdot \kappa \nabla \beta + \frac{\Lambda_D \Lambda_B}{9} \nabla \cdot (\nabla^2 \mathbf{q} + 2 \nabla (\nabla \cdot \mathbf{q})). \]  

(4.8)

The last two terms in the RHS of Eq. (4.8) become equal after taking the divergence: the first term can be written as

\[ \nabla \cdot \nabla^2 \mathbf{q} = \sum_{k=1}^{3} \frac{\partial}{\partial x_k} \left( \sum_{j=1}^{3} \frac{\partial^2 q_k}{\partial x_j^2} \right) = \sum_{k=1}^{3} \sum_{j=1}^{3} \frac{\partial q_k}{\partial x_k x_j^2}; \]  

(4.9)

while the second one becomes

\[ \nabla \cdot \nabla (\nabla \cdot \mathbf{q}) = \sum_{j=1}^{3} \frac{\partial^2}{\partial x_j^2} \left( \sum_{k=1}^{3} \frac{\partial q_k}{\partial x_k} \right) = \sum_{k=1}^{3} \sum_{j=1}^{3} \frac{\partial q_k^3}{\partial x_k x_j^2}; \]  

(4.10)

Equation (2.11) can thus be expressed as

\[ \nabla \cdot \mathbf{q} = -\nabla \cdot \kappa \nabla \beta + \frac{\Lambda_D \Lambda_B}{3} \nabla \cdot (\nabla (\nabla \cdot \mathbf{q})). \]  

(4.11)
Plug Eq. (2.10) into Eq. (4.11) to yield

\[
\frac{\partial \beta}{\partial t} = c_v \kappa \nabla^2 \beta + \frac{\Lambda D \Lambda_B}{3} c_v \frac{\partial}{\partial t} (\nabla^2 \beta) + Q - \frac{\Lambda D \Lambda_B}{3} \nabla^2 Q.
\] (4.12)

The Laplacian of source term in Eq. (4.12) only modifies its amplitude since a form of Eq. (4.7) is assumed. This term, however, may intensify the inhomogeneity of source term in other cases, calls for further study regarding its mechanism and impact. After performing Fourier transform in time and space on both sides of Eq. (4.12), the solution in frequency domain can be written as follows

\[
\Delta \tilde{\beta}(\eta, \xi_x, \xi_y) = \frac{\tilde{Q}(\eta, \xi_x, \xi_y)}{(iC_v \eta + \kappa_{\text{eff}}(\xi)^2)},
\] (4.13)

where \(\eta, \xi_x, \xi_y\) are transform variables corresponding to \(t, x, y\) respectively, \(\tilde{Q}(\eta, \xi_x, \xi_y) = 4\pi Q_0 \delta(\xi_x - \lambda)\beta_1 \beta_2 / (\beta_1^2 + \xi_y^2)\) is the Fourier transform of \(Q(t, x, y)\) in Eq. (4.7). The result is denoted with a \(\Delta\) because we consider the temperature change compared to the initial temperature (room temperature 295 K). In this equation, \(\kappa_{\text{eff}}(\xi)\) is the ETC given by

\[
\kappa_{\text{eff}}(\xi) = \kappa_{\text{bulk}} / (1 + \frac{\Lambda D \Lambda_B}{3} \xi^2),
\] (4.14)

\(\xi^2 = \xi_x^2 + \xi_y^2\) is the square of magnitude of spatial frequency.

According to Minnich [41], what is measured in 2D TTG experiment is the optical signal \(H(t, x)\) given by the following correlation

\[
H(t, x) = \int_0^\infty \Delta \beta(t, x, y) \exp(-\beta_2 y) dy,
\] (4.15)

where \(\beta_2\) is the absorption coefficient of the probe laser in the material. After taking Fourier transform of Eq. (4.15), the detected signal in the frequency domain is expressed as

\[
\tilde{H}(\eta, \xi_x) = \int_0^\infty \Delta \tilde{\beta}(\eta, \xi_x, \xi_y) \frac{2\beta_2}{\beta_2^2 + \xi_y^2} d\xi_y,
\] (4.16)

Substitution of Eq. (4.13) into Eq. (4.16) followed by an inverse Fourier transform in both time and space yields the following expression for the signal in 2D TTG experiments

\[
H(t, x) = \frac{4Q_0 \beta_1 \beta_2 c_v \xi_x}{c_v} \int_0^\infty \frac{\exp(-\kappa_{\text{eff}}(\xi)^2 t / c_v)}{(\beta_1^2 + \xi_y^2)(\beta_2^2 + \xi_y^2)} d\xi_y,
\] (4.17)

where \(\xi_x = \frac{2\pi}{P}\). In sum, with appropriate material properties given, the measured signal can be successfully constructed by Eq. (4.17).

### 4.2.3 Definition of \(\zeta\) in 2D

In 2D TTG, there is no explicit expression of the ETC \(\kappa_{\text{eff}}\). As shown in Eq. (4.14), the ETC \(\kappa_{\text{eff}}\) in 2D TTG depends on \(\xi\), which contains the integral variable in Eq. (4.17) for the signal. Although the signal calculated by Eq. (4.17) in the in-plane direction still maintains a sinusoidal
shape, it is now determined by a weighted integral of the temperature profile over the spatial frequency in the cross-plane direction. Consequently, the signal loses the single exponential behavior, and thus the explicit expression of ETC.

This is also the reason for a seemingly anisotropic ETC fitted in this experiment, as indicated by Minnich [41]. Minnich showed that the signals can also be fitted by setting anisotropic ETC $\kappa_x$ and $\kappa_y$, which unfortunately conflicts against the fact that silicon is thermally isotropic as a cubic crystal.

Although there is no explicit expression of the ETC, the nondimensional parameter $\zeta$, if extended to 2D situations, can still offer an estimation of the applicability of Fourier’s law. Considering that size-effects may also occur due to the shrinking size in the cross-plane direction, the characteristic length $L_c$ used for calculating the nondimensional parameter $\zeta$ is defined as

$$\frac{1}{L_c^2} = \frac{1}{L_{c,x}^2} + \frac{1}{L_{c,y}^2}. \quad (4.18)$$

Thus, we introduce the characteristic length in the cross-plane direction to the overall system characteristic length. Accordingly, we have

$$\zeta = \zeta_x + \zeta_y = \Lambda_D \Lambda_B \frac{1}{3L_{c,x}^2} + \Lambda_D \Lambda_B \frac{1}{3L_{c,y}^2} = \frac{\Lambda_D \Lambda_B}{3(\beta_1^2/\pi^2)} + \Lambda_D \Lambda_B \frac{1}{3(1/\beta_1)^2}. \quad (4.19)$$

where $L_{c,x}$ and $L_{c,y}$ are the characteristic length in $x$ (in-plane) direction and $y$ (cross-plane) direction, respectively. For example, in 1D TTG, the characteristic length in the cross-plane direction could be selected as the thickness of the thin film, if considered in a 2D situation. Thus, a qualitative estimation of the ETC for a silicon thin film of 400-nm thickness could be given as

$$\kappa_{\text{eff}} = \frac{\kappa_{\text{bulk}}}{1 + \zeta_x + \zeta_y} = \frac{\kappa_{\text{bulk}}}{1 + \zeta_y} \approx 91.45 \text{ W/mK}, \quad (4.20)$$

where $\zeta_x = 0$ if uniform heating is applied. This number is close to the value obtained by the measurement on that silicon thin film ($\kappa_{\text{diff}} = 92 \text{ W/mK}$). However, it should be noted that this calculation cannot provide a quantitative description of ETC due to the reasons mentioned before, nor can it include the effects from the boundary, which is generally important or even dominant in thin film cases. This example is only to show the $\zeta$ in 2D situations. Similarly, a $\zeta_z$ can be defined to introduce another component to $\zeta$ so that it also works for 3D situations. Hence, $\zeta$ has been successfully extended to multidimensional situations.

### 4.3 Validations and predictions for 2D TTG

To validate the results by the TPHC model, the published results by both experiments and theoretical work are compared with our calculations. In addition, the nondimensional parameter $\zeta$ is also illustrated to show a quick evaluation on the applicability of Fourier’s law.

#### 4.3.1 Comparison with experiments

As validation, the analytical solution of the signal is first compared to the published experimental results by Johnson et al. [37,40] in Fig. 4.4. The thermophysical properties of bulk GaAs
Figure 4.4: Comparison between analytical solution by the TPHC model and experimental results: (a) by Johnson et al. [40] with the thermal grating periods are $P = 10 \mu m$, 4.9 $\mu m$, 3.2 $\mu m$, 2.05 $\mu m$ from high to low, and (b) by Johnson et al. [37] with the thermal grating periods are $P = 4.9 \mu m$, 3.7 $\mu m$, 2.8 $\mu m$, 2.4 $\mu m$ from high to low.

Table 4.1: Thermophysical properties of materials used in 2D TTG study ($\beta_2 = \beta_1$) for the TPHC model

<table>
<thead>
<tr>
<th>Material</th>
<th>$c_p$ (J/m$^3$K)</th>
<th>$v$ (m/s)</th>
<th>$\kappa_{\text{bulk}}$ (W/mK)</th>
<th>$\Lambda_B$ ($\mu m$)</th>
<th>$\beta_1$ (nm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicon</td>
<td>$1.66 \times 10^6$</td>
<td>6400</td>
<td>148</td>
<td>7.1</td>
<td>1/580</td>
</tr>
<tr>
<td>GaAs</td>
<td>$1.75 \times 10^6$</td>
<td>3700</td>
<td>45</td>
<td>10</td>
<td>1/150</td>
</tr>
<tr>
<td>Sapphire</td>
<td>$2.63 \times 10^6$</td>
<td>7766.9</td>
<td>41.13</td>
<td>1.6</td>
<td>N/A</td>
</tr>
</tbody>
</table>

used in the simulation are summarized in Table 4.1. Specifically, the thermal conductivity of bulk GaAs is given as $\kappa_{\text{bulk}} = 45$ W/mK. Due to the missing of an explicit expression to extract $\Lambda_B$, we need to determine it by fitting one set of the 2D TTG experimental data. Here we choose a case $P = 2.05 \mu m$ where the nondiffusive effects are most significant, and find good match when $\Lambda_B = 10 \mu m$. With this fitted $\Lambda_B$ for bulk GaAs, we demonstrate that the TPHC model can be used to predict the thermal decay curve for all other grating periods. The predictions are compared with the experimental results in Fig. 4.4. Further studies are required for more accurate measurements of the ballistic transport length $\Lambda_B$ in bulk GaAs.

For the case of $P = 2.05 \mu m$, the results by Fourier’s law using Eq. (4.17) with constant apparent thermal conductivity $\kappa = 45$ W/mK is also plotted in Fig. 4.4(a) and (b) for comparison. These two figures show that the Fourier’s law remarkably overpredicts the thermal grating decay. This overprediction becomes more and more significant as the grating period $P$ decreases. Moreover, for large grating period where $P = 10 \mu m$ and $\zeta_x = 0.0274$, it is still found to deviate from the experimental result (the small window of Fig. 4.4(a)). Although $\zeta_x$ is just slightly larger than the critical value $\zeta_c = 0.01$ given in the 1D TTG part, the discrepancy between the two simulated curves is not expected to be so significant. This discrepancy is caused by the nondiffusive thermal transport in the cross-plane direction, as $\zeta_y = 2.71$ is calculated with the penetration depth of the pump laser.
$\beta_1^{-1}$ as the characteristic length in this direction, implying that the discrepancy will still exist for even larger $P$. On the other hand, the gap between the results by the TPHC model and Fourier’s law for $P = 2.05\mu m$ is simultaneously contributed by the nondiffusive effects in both in-plane and cross-plane directions.

Overall, there is good agreement between predictions based on the TPHC models and experiments, which demonstrates that the TPHC model is able to capture the nondiffusive heat transfer in 2D TTG experiments. In addition, the failure of Fourier’s law is also identified as predicted by the nondimensional parameter $\zeta$, suggesting a successful extension of $\zeta$ into 2D situations.

4.3.2 Comparison with spectral BTE

In addition to the experimental results shown in the previous subsection, Minnich [41] constructed synthesized theoretical results using full-spectral BTE with the phonon dispersion obtained by \textit{ab initio} simulation package. In our calculation, all the needed material properties of bulk silicon are the same as in the 1D TTG part, except for the thermal conductivity $\kappa = 148$ W/mK for bulk silicon. $\Lambda_B$ here is obtained from 1D TTG as $\Lambda_B = 7.1\mu m$. On the other hand, the penetration depth of pump and probe have to be specified here. However, this 2D TTG case, unfortunately, has not be experimentally reported, possibly due to the transparency of silicon to the laser pump and probe generally in use. Although such a transparency leaves the penetration depth unknown, it implies a very large value of penetration depth (i.e. tens of $\mu m$). Here, the absorption coefficients for pump and probe are set to be equal ($\beta_1 = \beta_2 = 1/580$ nm$^{-1}$) for the TPHC model.

Figure 4.5(a) shows the comparison between the results from the TPHC model and full-spectral BTE by Minnich [41]. Since the penetration depths for both pump and probe are small (i.e., less than 1 $\mu m$), the agreement between the nondiffusive models and Fourier’s law is not expected.
even for the largest grating period. Furthermore, Fig. 4.5(b) shows artificial cases with even smaller grating period to contrast the TPHC model and Fourier’s law. This figure also indicates that the overprediction on thermal decay rate by Fourier’s law deviates monotonically away from the TPHC model. This can be explained by the failure of the diffusive assumption on which Fourier’s law is based, as the thermal transport behaves nondiffusively or even ballistically in such a length scale. Similar trend has been observed in 1D TTG results.

An interesting implication from the 2D TTG analytical solutions is the interchangeability of $\beta_1$ and $\beta_2$. One may already notice that based on Eq. (4.17), identical result can be obtained even if $\beta_1$ and $\beta_2$ are switched. Physically, this indicates that, either the pump laser or the probe laser may result in the nondiffusive effects of the signal. In other words, the nondiffusive phenomena revealed by this experimental technique are probe-dependent, if assuming Eq. (4.17) is physically correct for the signal. But this hypothesis needs further experimental validation.

### 4.3.3 Impact of cross-plane nondiffusive effects

As shown in the previous section, in neither GaAs cases nor silicon cases is the diffusive limit for bulk material reached. However, the diffusive limit is indispensable for the study of nondimensional parameter $\zeta$ as a criterion. Although this criterion can be predicted by the TPHC model, a experimental validation is still desirable as solid evidence. Fortunately, the diffusive limit for bulk sapphire is known from MG-X-ray study that will be shown later in this chapter. We put it here to illustrate the criterion for diffusive limit. Specifically, in the case where the grating period $P = 3.24 \mu m$, there is good match between the optical signal and Fourier’s law prediction, implying that the diffusive limit has been reached around $P = 3.24 \mu m$. In other words, the TPHC model and Fourier’s law should agree with each other with $P = 3.24 \mu m$.

All the needed material properties of sapphire are specified in Table 4.1 ($\Lambda_B = 1.6 \mu m$ is fitted for sapphire in the next section), except for the penetration depth of the pump and probe. Due to the transparency of sapphire, no 2D TTG experiment has been reported on this material, just as the silicon. Similar to the silicon case, the penetration depths for pump and probe are assumed large and set to be equal so that $\beta_1 = \beta_2 = 0.5 \mu m$, as in previous studies [41]. We later will show the effects from this value. Based on this number, Fig. 4.6(a) shows the result.

In this figure, the grating periods are the same as selected in MG-X-ray case for later comparison regarding the size-dependence of interface resistivity. The signals by Fourier’s law (dashed lines) are compared with the results by the TPHC model (solid lines). Still, with decreasing thermal grating period, there is a faster decay of the normalized optical signal. Remarkable difference in the predictions between the TPHC model and Fourier’s law is also observed for two smallest cases with $P = 0.32 \mu m$ and $0.76 \mu m$, as in both GaAs and silicon cases. To match the results of the TPHC model, the ETC has to be used in Fourier’s law. With increasing thermal grating period $P$, the difference in predictions between the TPHC model and Fourier’s law decreases. When the thermal grating period increases to $P = 3.24 \mu m$, the difference in predictions between the TPHC model and Fourier’s becomes negligible. In such case where the nondimensional parameter $\zeta$ is expected to be small, it can be calculated as $\zeta = 8.5 \times 10^{-3} < 0.01$, thus validating our previous assumption.

On the other hand, to study the influence by cross-plane nondiffusive effects, $\zeta_y$ is calculated with characteristic length to be penetration depth of pump $\beta_1^{-1}$. Parametric study on this parameter has been done and shown in Fig. 4.6(b). In this study, the signal itself is sensitive to $\zeta_y$ in the range of interest, but we notice that the difference between the signals calculated by two models
remains unchanged within certain region. For example, in $P = 3.24 \mu m$ case, the signals calculated by the TPHC model and Fourier’s law are always the same if $\zeta_y$ is not very large. Given that it is the difference that describes the nondiffusive heat transfer, we consider a quantitative description of this difference necessary. To quantify this difference, we apply a simple definition to describe how different two curves are. Eq. 4.21 is used to calculate the average difference $\delta_{AD}$ between two curves.

$$\delta_{AD} = \frac{1}{n} \sum_{k=1}^{n} |1 - \frac{H_F(t_k, x = 0)}{H_T(t_k, x = 0)}|,$$  \hspace{1cm} (4.21)$$

where the subscript $F$ represents Fourier’s law, $T$ represents the TPHC model. Ideally, this average difference $\delta_{AD}$ is 0 if two curves are identical, and its maximum is 1 if both $H_F$ and $H_T$ are always positive. However, it is approaching 0 if the tails of two calculated signals are considered. Consequently, we choose a criterion 0.01. That is, we only consider the part of the curve where the signal is larger than 0.01 and use this segment to calculate the average difference $\delta_{AD}$. On the other hand, this difference also depends on the number of the discretized time steps selected for calculation, or equivalently, resolution. We have already tested different resolutions to assure that the result is independent of resolution.

As shown in Fig. 4.6(b), in a wide range of $\zeta_y$, the average difference $\delta_{AD}$ remains unchanged for all four cases, indicating that in this given range, the nondiffusive effects in the cross-plane heat transfer is not significant in overall nondiffusive effects. However, $\zeta_y$ weighs more in the overall $\zeta = \zeta_x + \zeta_y$ when it approaches the value of $\zeta_x$, and this results in the increase of $\delta_{AD}$, as observed in the GaAs case with $P = 10 \mu m$. This component becomes dominant when the penetration depth becomes extremely small.

Qualitatively, four different regimes can be divided in Fig. 4.6(b). The left bottom corner regime represents diffusive regime where the nondiffusive effect from both in-plane and cross-plane

Figure 4.6: (a). Comparison of temperature decay curves in 2D TTG in sapphire for four different grating periods between analytical solutions of the TPHC model (solid lines) and Fourier’s law results (dashed lines); (b). The average difference $\delta_{AD}$ calculated by Eq. (4.21) with varying $\zeta_y$. 

As shown in Fig. 4.6(b), in a wide range of $\zeta_y$, the average difference $\delta_{AD}$ remains unchanged for all four cases, indicating that in this given range, the nondiffusive effects in the cross-plane heat transfer is not significant in overall nondiffusive effects. However, $\zeta_y$ weighs more in the overall $\zeta = \zeta_x + \zeta_y$ when it approaches the value of $\zeta_x$, and this results in the increase of $\delta_{AD}$, as observed in the GaAs case with $P = 10 \mu m$. This component becomes dominant when the penetration depth becomes extremely small.

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Qualitatively, four different regimes can be divided in Fig. 4.6(b). The left bottom corner regime represents diffusive regime where the nondiffusive effect from both in-plane and cross-plane
directions are negligible. The left top corner regime marks the cases with strong nondiffusive effects from the in-plane direction and negligible effects from the cross-plane direction (i.e., 1D TTG case). Analogically, the right bottom corner represents the cross-plane nondiffusive dominant case (i.e., cross-plane ETC study), and the right top corner stands for the strong nondiffusive regime. Hence, the nondiffusive effects from either direction will lead to the failure of Fourier’s law, but this failure can be predicted by $\zeta$ on the corresponding direction, which shows potential to assist the engineering design for a more general multidimensional situation.

However, the strong nondiffusive regime is out of the reach of the TTG experiments. As given in Eq. (4.1), the thermal grating period is limited by the wavelength of the heating laser, which typically is hundreds of nanometers. To reach a even smaller grating size, one possible solution is to apply the electron beam heating. However, so far there is no such result report. Siemens et al. [42] offered an alternative way to further shrink the grating size by the implementation of the metallic grating.

### 4.4 Applications of the TPHC model on the MG-X-ray experiments

In previous sections, we focused on the TTG experiments where the thermal interface is avoided. The thermal interface, however, is ubiquitous and playing an important role in lots of applications. It is also reported to behave differently from the prediction by diffusive theory in certain length scales, and is studied by different nanothermometry techniques. Among these techniques, Time-domain thermoreflectance (TDTR) [43–45], for example, is an established experimental technique contributing to the discovery of the size-dependence of the interface resistivity or it reciprocal, conductance [43, 46, 47].

To further study the thermal transport in a smaller scale than the TTG experiment can reach, and to study the size-dependence of the interface resistivity, Siemens et al. designed a new experimental technique. They applied nanoscale heating sources using a technique of laser absorption of metallic grating (MG), and measured the thermal decay of the metallic grating using diffracted signal of soft X-ray [42]. Through this technique, they reported the first observation and quantitative measurement of transition from diffusive to quasi-ballistic thermal transport [42].

Currently, though Fourier’s law is still used in the data analysis of the MG-X-ray experiment, there should be both size-dependent ETC and size-dependent thermal interface resistivity in the MG-X-ray experiments that cannot be described by Fourier’s law. Moreover, it is infeasible to determine both ETC and thermal interface resistivity simultaneously in the MG-X-ray experiments based on Fourier’s law. Consequently, in the data analysis of the MG-X-ray experiments [42, 48], thermal conductivity of the substrate was fixed as a constant so that the only size-dependent parameter is the thermal interface resistivity. This size-dependent interface resistivity was determined from thermal decay curves for different sizes of metallic gratings, but it actually also carries the size-dependence of the ETC in addition to the size-dependence of its own. To obtain the individual size-dependence of the interface resistivity, it is necessary to apply high-fidelity nondiffusive heat transfer model in the MG-X-ray data analysis that can take care of the size-dependence of the ETC. Unfortunately, so far to the best of our knowledge, these results on the MG-X-ray experiments have not been reproduced by any other existing nondiffusive heat transfer model.

Here, we apply the TPHC model to study the size-dependent nondiffusive heat transfer both inside the substrate and across the interface in smaller nano-sized gratings compared to the TTG.
Analytical solutions as implemented in the TTG experiments unfortunately are not available in this case. Thus, the MG-X-ray experiments are studied by numerical approaches and the results are compared with the experiments by Siemens et al. [42] as validations. In addition to the size-dependence of ETC, the size-dependence of interface resistivity is also indirectly extracted, suggesting the potential of the TPHC model to explore the nondiffusive thermal transport across the interfaces.

4.4.1 Physical situations of the MG-X-ray experiment

![Figure 4.7: (a) Schematic of metallic gratings for the measurement of nondiffusive heat transfer; (b) Computational domain for one period of metallic grating.](image)

Fig. 4.7(a) shows the schematic of the MG-X-ray experiments in which the nano-sized heaters are periodic nickel stripees with thickness of 20 nm, length of 120 µm and various widths [42]. These nickel stripes are attached on the top surface of a sapphire substrate. Here, the thickness of the substrate is several orders of magnitude larger than that of the nickel stripes so that the substrate can be treated as a semi-infinite body. Considering that the length of the heaters is at least two orders of magnitude larger than their width and thickness, heat transfer in MG-X-ray experiments can be simplified as 2D. Moreover, because the laser beam diameter (∼ 100 µm) is much larger than the spatial period of MGs, one typical MG period is representative and selected as the computational domain shown in Fig. 4.7(b). Here, the in-plane direction along the width of the MG is chosen to be x direction and the cross-plane direction is denoted as y direction.

When the substrate and nickel gratings are exposed to an ultrashort (∼ 25 fs) pulse of laser pump at a wavelength of 800 nm, only the nickel gratings are heated because of the transparency of the sapphire substrate to the laser. After the ultrashort laser heating, electrons in nickel gratings are excited first and passing their thermal energy to phonons in the nickel heaters through electron-phonon interactions. In less than 10 ps, electrons and phonons can reach local thermal equilibrium. Compared with the whole thermal decay process (several nanoseconds) in the MG-X-ray experiments, the electron-phonon equilibration process in nickel gratings is so short that Fourier’s law is applicable for the heat conduction in the metal heaters. In the substrate, both the TPHC model and Fourier’s law will be used to calculate thermal transport and compared with each other. Since the model and the numerical schemes have been provided in Chapter 2 and 3, respectively, only a brief summary of the boundary and initial conditions for MG-X-ray is given below for brevity.
4.4.1.1 Boundary & initial conditions

In 2D Cartesian coordinate shown in Fig. 4.7(b), the computational domain is composed of two subdomains: the nickel heater and the sapphire substrate. In the heater subdomain, all boundaries except for the interface are insulated. Since Fourier’s law is implemented in this subdomain, the boundary conditions can be given as

\[-\kappa_h \frac{\partial T}{\partial n} = \begin{cases} \frac{T_h - \beta}{r}, & \text{on heater-substrate interface,} \\ 0, & \text{else.} \end{cases}\]  

(4.22)

where the subscript \( h \) represents the heater subdomain, while the physical quantity \( \beta \) is the nonequilibrium absolute temperature of the subdomain. In this equation, \( T_h \) and \( \beta \) are at the same location.

For the substrate subdomain, while the equation describing heat flux \( \vec{q} \) in Fourier’s law is a first-order partial differential equation (PDE), Eq. (2.11) in the TPHC model is a second-order PDE. Therefore, one more boundary condition than Fourier’s law is required for the TPHC model. As shown in Fig. 4.7(b), periodic boundary condition is applied in \( x \) direction for both \( q_x \) and \( q_y \) of the substrate subdomain. In \( y \) direction, \( \frac{\partial q_x}{\partial y} = 0 \) is applied at boundary \( y = 0 \) and \( y = y_{\text{max}} \to \infty \). On the top surface of the substrate, the boundary condition for the normal flux is similar to Eq. (4.22), which can be specified as

\[ q_y = \begin{cases} \frac{T_h - \beta}{r}, & \text{on heater-substrate interface,} \\ 0, & \text{else.} \end{cases}\]  

(4.23)

where \( T_h \) and \( \beta \) represent the temperature of nickel stripe heater and the nonequilibrium temperature of the substrate at the interface, respectively, \( r \) is the thermal interface resistivity [42]. The bottom boundary of the substrate is insulated.

It should be noted that there are two singularity points located at the two corners of the heater-substrate interface (see Fig. 4.7(b)). To eliminate the discontinuity which may cause numerical issues, the heat flux across the interface is smoothed to be an error function shape profile with the total energy conserved during the simulation process. The effects from this treatment are examined and show negligible influence based on our numerical experiments.

As the initial condition, the normalized temperatures in the nickel heater and the substrate are specified as \( T_h = 1 \), and \( \beta = 0 \), respectively.

4.4.1.2 Signal model

In the MG-X-ray experiments, a signal model is required to link the calculated temperature field to the dynamic soft X-ray diffraction signal [42]. In Ref. [42, 48], a Fresnel optical propagation model was applied to link the soft X-ray diffraction signal to the deformation of the grating system surface. The deformation of the grating surface consists of two components: (1) periodic surface acoustic waves (SAWs) induced by the deformation of the metallic grating that oscillates with damping, and (2) the thermal expansion of the system that decays monotonically with time. In these two components, it is the thermal expansion of the system that represents the cooling dynamics of the nickel nanostructures [42]. Thus, the thermal expansion, or deformation, of the system is computed based on the transient temperature field from the TPHC model, and the SAW component is fitted by a sinusoidal oscillating term with an exponentially decayed amplitude. These two com-
ponents are superimposed. The signal model to link the surface deformation and the temperature field is briefly described below.

Considering the relatively small thickness and volume of the nickel heater, its thermal expansion is approximately uniform. The averaged thermal expansion of the nickel heater is described by

$$\Delta h_h(x) = \alpha_h \Delta \bar{T}_h(x), \quad (4.24)$$

where $\alpha_h$ is the coefficient of linear thermal expansion of the nickel heater, and $\bar{T}_h$ is the average temperature (over $y$ direction) of the nickel heater at corresponding $x$ position. Meanwhile, the thermal expansion of the substrate surface is given by

$$\Delta h'_s(x) = \frac{2(1+\nu_s)\alpha_s}{3\pi} \int_{x_1} \int_{y} \beta(x_1, y) \frac{yd_{x_1}dy}{(x-x_1)^2 + y^2}, \quad (4.25)$$

where $\alpha_s$ is the coefficient of linear thermal expansion of the substrate, and $\nu_s$ is the Poisson’s ratio of the substrate. This formula is for the thermal expansion in a semi-infinite body [49] with a single source, but it is also applicable in this case because the resultant deformation at one position can be considered as a superposition of the deformation due to each of the heaters when they individually show up. Consequently, the integration in Eq. (4.25) is carried out for multiple periods of gratings and the results are superimposed to obtain the thermal expansion of the system:

$$\Delta h_s(x) = \sum_{j=-\infty}^{\infty} h'_s(x + jP), \quad (4.26)$$

Typically, considering only two neighboring periods is enough, so that the $j$ in Eq. (4.26) is from -2 to 2.

It should be noted that the detected diffraction signal in the experiments indicates the relative surface deformation rather than the absolute value of surface displacement. For example, the diffraction signal won’t change for uniform surface deformation in $y$ direction. As shown in Fig. 4.7(b), the deformation at point A (peak deformation) is a superposition of two parts: the deformation of the heater and the deformation of the substrate; while the deformation at point B (valley deformation) is just the local deformation of the substrate. Therefore, the difference between the calculated deformation of point A and point B represents the overall relative surface deformation $\Delta h$

$$\Delta h = (\Delta h_{h,A} + \Delta h_{s,A}) - \Delta h_{s,B}. \quad (4.27)$$

The relative deformation $\Delta h$ is normalized before it is compared with optical signals in MG-X-ray experiments.

### 4.4.2 Numerical solutions of MG-X-ray

As specified in Chapter 3, finite difference method is implemented for numerically solving the TPHC model. The solution by Fourier’s law can be obtained by setting $\Lambda_B = 0$ in the solution of the TPHC model. For brevity, the discretization process is not shown here. The solutions from both models are directly given and compared with experimental results by Siemens et al. [42].
First, we want to address some issues with Fourier’s law.

4.4.2.1 Uncertainty of data analysis in MG-X-ray using Fourier’s law

Currently, modified Fourier’s law with a size-dependent ETC is still widely used in the data analysis of pump-probe nanothermometry. If the modified Fourier’s law is applied for the data analysis of MG-X-ray experiments, size-dependent ETC should be used, as indicated in TTG part. Thus, the unknown size-dependent ETC needs to be determined from the experimental results. In addition to the ETC, there is another unknown parameter, the size-dependent thermal interface resistivity \( r \), which also has to be determined from the MG-X-ray experiments [42]. Accordingly, two constraints are needed for each grating period. However, there is only one output for the MG-X-ray experiments, that is the diffracted signal. This is still possible in some special cases, for example, the two parameters influence the output in different time scales. Unfortunately, this is not the case in MG-X-ray because similar impacts on the response of thermal decay signal due to the change of the ETC and \( r \) are observed, indicating that different combinations of the ETC and \( r \) may fit the same thermal decay signal. Therefore, it is almost impossible to extract both ETC and \( r \) simultaneously from MG-X-ray experiments.

As an example, Fig. 4.8 shows the comparison of two calculated signal traces with different combinations of ETC and \( r \) using Fourier’s law. In this case, the linewidth of the stripe heater on the sapphire substrate is \( L = 80 \) nm. The solid line is the experimental signal plotted as reference. Dashed line represents for the predictions with the thermal conductivity as the bulk value \( \kappa_{\text{bulk}} = 41.13 \) W/m\( ^2 \)K, and \( r = 4.4 \times 10^{-9} \) Km\( ^2 \)/W. The dashed-dot line shows the predictions with ETC \( \kappa_{\text{eff}} = 0.53\kappa_{\text{bulk}} \), and \( r = 2.0 \times 10^{-9} \) Km\( ^2 \)/W. Both predictions are able to fit the experimental signal. In fact, for the same experiment, there are infinite combinations of the ETC and \( r \) that can fit the experimental data. Consequently, Fourier’s law fails to provide an accurate physical interpretation to the MG-X-ray experiments. In order to reveal the mechanisms of the nondiffusive heat transfer in the MG-X-ray experiments, we apply the TPHC model to reinterpret the experimental data.

4.4.2.2 Determination of \( \Lambda_B \) in the TPHC model

There are two independent parameters in the TPHC model: thermal conductivity \( \kappa_{\text{bulk}} \) and ballistic phonon MFP \( \Lambda_B \). The thermal conductivity of bulk sapphire material [42] is given as \( \kappa = 41.13 \) W/m\( ^2 \)K. Here we omit the subscript since from now on, \( \kappa \) without subscript unambiguously means the thermal conductivity of the corresponding bulk material. Other material properties for bulk sapphire are given in Table 4.1. Mechanical properties are: Poisson ratio \( \nu_s = 0.25 \), linear coefficient of thermal expansion for the nickel heater \( \alpha_h = 12.77 \times 10^{-6} \) K\(^{-1} \) and substrate \( \alpha_s = 5.31 \times 10^{-6} \) K\(^{-1} \).

The value of \( \Lambda_B \) is determined through data fitting based on experimental results by Siemens et al. [42]. In the experiments, the width of nickel heater (\( L \)) ranges from 80 to 810 nm. It was found that there is increasing nondiffusive heat transfer with decreasing heater width \( L \). For \( L = 80 \) nm, the nondiffusive effects are most significant compared with other cases. Therefore, we use the thermal decay data of experiment with \( L = 80 \) nm to determine the value of \( \Lambda_B \).

Although there is only one constraint for extracting two parameters, it is still possible according to the sensitivity study. In the sensitivity study, the signal with \( \Lambda_B = 1.6 \) µm and \( r = 2.9 \times 10^{-9} \) Km\( ^2 \)/W calculated by Eq. (4.27) is plotted as the reference. To study the sensitivity of...
thermal decay curve to the changes of two parameters, we either fix $\Lambda_B = 1.6\, \mu m$ and change $r$ by $\pm 20\%$ around $r = 2.9 \times 10^{-9}\, Km^2/W$, or fix $r = 2.9 \times 10^{-9}\, Km^2/W$ and change $\Lambda_B$ by $\pm 50\%$ around $\Lambda_B = 1.6\, \mu m$. These results are also plotted in Fig. 4.9. It can be seen that the signal is not sensitive to $\Lambda_B$ within roughly 500 ps; while after 500 ps, it becomes sensitive to $\Lambda_B$. This study suggests that $r$ can be fitted first using the signal within 500 ps. Afterwards, the value of $\Lambda_B$ is the only unknown parameter to be determined.

Accordingly, the fitting strategy can be briefly described here. At first, the thermal decay curve is extracted from experimental data by filtering out the oscillatory SAW component in the signal. Second, a least square method provided in MATLAB curve fitting toolbox is used to simultaneously extract the two parameters ($\Lambda_B$ and $r$) by fitting the thermal decay curve of the MG-X-ray experiment with $L = 80\, nm$. The reference curve shown in Fig. 4.9 is the best fitting result where $\Lambda_B = 1.6\, \mu m$ and $r = 2.9 \times 10^{-9}\, Km^2/W$. Even though there exists high uncertainty (about 50%) in the value of $\Lambda_B = 1.6\, \mu m$, the TPHC model can take the size-dependent nondiffusive effects into account. Thus, we expect that the TPHC model will provide more reliable results on the thermal interface resistivity $r$ than Fourier’s law in the data analysis of MG-X-ray experiments.

### 4.4.2.3 Reinterpretation of MG-X-ray experiments on sapphire

Here we use the TPHC model with $\Lambda_B = 1.6\, \mu m$ to reinterpret the MG-X-ray experiments by Siemens et al. [42] first. Overall, there are four different cases with the nickel heater width $L = 810, 350, 190$ and $80\, nm$, respectively. To extract the value of $r$ from experimental data, the fitting process is the similar to the one used for extracting $\Lambda_B$, except that now $r$ is the only fitting parameter. After the only fitted parameter $r$ is known, the signal is recalculated by the TPHC model and the SAW component is superimposed on top of the thermal decay as a sinusoidal oscillating term with an exponentially decayed amplitude. The data fitting results using the TPHC model are compared with experiments in Fig. 4.10. The data fitting results using Fourier’s law with
Figure 4.9: Sensitivity of the thermal decay curve for $L = 80$ nm to the change of thermal interface resistivity ($r$) and nondiffusive phonon MFP ($\Lambda_B$).

\[ \kappa = 41.13 \text{ W/m}^2\text{K} \] are also plotted in the figures for comparison. Although for all four different cases, the fitting results from both the TPHC model and Fourier’s law agree with the experimental signals very well, the improvement by the TPHC model lies in extracted $r$.

Through data fitting, the value of thermal interface resistivity $r$ for four different cases are fitted and summarized in Table 4.2 with the unit $10^{-9}$ Km$^2$/W. The results of $r$ vs. $L$ are also plotted in Fig. 4.11. It is found that there is monotonic increase of thermal interface resistivity with decreasing linewidth of the nickel heater $L$ based on the results of Fourier’s law. Compared with the results of Fourier’s law, there is faster increase of the thermal interface resistivity with the decreasing $L$ when $L > 190$ nm. When $L$ decreases further to 80 nm, the thermal interface resistivity extracted by the THPC model decreases.

Due to the monotonic behavior of the size-dependence of ETC, such monotonic behavior is also expected though without any evidence. However, as the characteristic length approaches the grating period, the nondiffusive heat transfer may go out of the applicable range of the TPHC model. As shown in Fig. 4.6(a), the substrate ETC in a similar periodic heating pattern (2D TTG) decreases monotonically. Similarly, the same trend is also expected here, as $\xi_x$ becomes larger and larger. The size-dependence of $r$ here will be further studied using the EG-BTE, which is still applicable for this length scale.

<table>
<thead>
<tr>
<th>$L$</th>
<th>810 nm</th>
<th>350 nm</th>
<th>190 nm</th>
<th>80 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_{Fourier}$</td>
<td>2.1</td>
<td>2.9</td>
<td>3.1</td>
<td>4.4</td>
</tr>
<tr>
<td>$r_{THPC}$</td>
<td>2.1</td>
<td>3.3</td>
<td>3.6</td>
<td>2.9</td>
</tr>
</tbody>
</table>

Table 4.2: Summary of fitted interface resistivity $r$ with different nickel heater width $L$ in sapphire cases. The unit of $r$ is $10^{-9}$ Km$^2$/W.
Figure 4.10: Comparison of the fitting results of TPHC model and Fourier’s law with the experimental data [42]. The unit of the thermal interface resistivity \( r \) is \( 10^{-9} \text{Km}^2/\text{W} \).

### 4.5 Discussion

In this section, we specify the limitations of the TPHC model, its range of applicability, and its problems.

**Limitations** The limitations of the TPHC model lie in the fact that this model is a phenomenological model and is unable to reveal the theoretical details about the phonon transport from a physical perspective. Its mathematical simplicity is obtained at the cost of losing information in physics, however, it aims at the experimental data analysis and engineering applications where the requirements for detailed physical characteristics are not so high.

**Range of applicability** The range of applicability of the TPHC model is the weak nondiffusive thermal transport regime. For example, in MG-X-ray experiments, the smallest grating period with \( L = 80 \text{nm} \) actually approaches the boundary of this range. Another example is the MG results on silicon substrate by Hoogeboom-Pot et al. [48]. They offered more experimental data with extremely small grating periods, but the TPHC model fails to extract a reliable result. Therefore, these experimental results will be analyzed by the EG-BTE in Chapter 5.
Problems The boundary condition is a problem for the TPHC model because it requires one more boundary condition than Fourier’s law. However, it is interesting to note that, based on the assumption that the heat flux is irrotational, the last two terms in Eq. (2.11) are exactly the same, as shown in Eq. (2.35). However, is the heat flux physically irrotational? So far there is no answer.

4.6 Summary

In this chapter, the Two-Parameter Heat Conduction (TPHC) model is applied on 1D/2D transient thermal grating (TTG) experiments and metallic grating experiments (MG-X-ray experiments).

In 1D TTG experiments, the analytical solution of the TPHC model is derived. From the analytical solution, explicit expressions of effective thermal conductivity (ETC) and the thermal decay rate are obtained and show excellent agreements with the experimental results. In addition, the nondimensional parameter $\zeta$ proposed in Chapter 2 also quantitatively predicts the change of the ETC or thermal decay rate due to the nondiffusive effects.

In 2D TTG experiments, the analytical solution of the TPHC model is also derived and explains the seemingly anisotropic ETC discovered in the experiments. The good matches between TPHC results and results by both experiments and full-spectral Boltzmann transport equation (BTE) marks the accuracy of the TPHC model in the 2D TTG experiment. On the other hand, $\zeta$ is extended to multidimensional situations and validated through the comparison with Fourier’s law.

In MG-X-ray experiments, the numerical solution of the TPHC model is obtained to replace Fourier’s law in the experimental data analysis. Multiple solutions by Fourier’s law reveal the challenges for Fourier’s law, while the TPHC model is able to capture the nondiffusive heat transfer inside the substrate and thus indirectly extract a clean size-dependence of the interface resistivity.
To sum up, the TPHC model is validated by multiple experiments, and proves its capability for the experimental data analysis as well as engineering applications. The nondimensional parameter $\zeta$ proposed in this thesis is also validated, and provides a quick evaluation of the applicability of Fourier’s law, if certain critical value is given according to the requirements of the engineering applications.
Chapter 5

Application of the Enhanced Gray Boltzmann Transport Equation

In this chapter, the Enhanced Gray Boltzmann transport equation (EG-BTE) is applied on multiple nondiffusive heat transfer experiments, including 1D/2D transient thermal grating (TTG) experiment, metallic grating experiment measured by diffracted X-ray (MG-X-ray experiment), and the thermal wave experiments at low temperature. Different from the its simplified version, the TPHC model, the EG-BTE in this thesis focuses on the fundamental study. In addition to the features of the nondiffusive heat transfer captured by the TPHC model, the EG-BTE explains a wide range of thermal transport covering all ballistic, transitional and diffusive regimes. Moreover, it also suggests the observation of the ballistic thermal transport at room temperature, which is a new step towards a better understanding of the physics behind heat conduction.

5.1 Applications of the EG-BTE on 1D TTG experiments

The experimental setup as well as the physical situation for the 1D TTG experiment has been given in Chapter 4. For brevity, we skip the repeating description and directly derive the analytical solution of 1D TTG based on the EG-BTE here.

5.1.1 Analytical solution by the EG-BTE

Since the physical situation of the 1D TTG experiment is still the same as in Chapter 4, the mathematical description of the 1D TTG experiment can still be given by the source term in Eq. (4.2). In this case, the system stays at equilibrium state initially, so that the initial conditions can be given as

\[
\begin{aligned}
    e(t = 0, x) &= 0, \\
    \frac{\partial e}{\partial t} \bigg|_{t=0} &= 0,
\end{aligned}
\]

if the initial phonon energy density is selected as the reference energy density.

However, there is an alternative way for modeling the physical situation in the 1D TTG
experiment. That is, to set the initial periodic temperature field so that the initial conditions are:
\[
\begin{align*}
  e(t = 0, x) &= e_0 \exp (i\lambda x), \\
  \left. \frac{\partial e}{\partial t} \right|_{t=0} &= 0.
\end{align*}
\] (5.2)

In this way, the source term is \( Q = 0 \). As explained in Chapter 4, we can set the magnitude \( e_0 = 0 \) because of the normalization of the final signal. By applying these initial conditions one presumes that the absorption of the laser heating is infinitely quick.

For the TPHC model and gray BTE, these two mathematical descriptions of the 1D TTG experiments are equivalent, because both of these two are first order in time. These two descriptions are also equivalent if the thermal decay time is long enough (i.e., longer than 20 ns).

On the other hand, the EG-BTE in 1D situation is expressed by Eq. (2.21) in a distribution function form. Here we follow the similar procedure as in Chapter 2 to convert it into an energy form:
\[
\tau_c \frac{\partial^2 e}{\partial t^2} + \frac{\partial e}{\partial t} + \mu v \frac{\partial e}{\partial x} = -\frac{e - e^0}{\tau_R} - \tau_c \mu v \frac{\partial^2 e}{\partial t \partial x} + \hat{Q},
\] (5.3)

where \( \mu = \cos \theta \) is the directional cosine.

The boundary conditions for this situation are periodic for both ends of the computation domain, as implemented in the TPHC model. With the governing equation and auxiliary conditions, here we derive the analytical solutions for both of the two descriptions. For brevity, we call the first option (with instantaneous source term and all zero initial conditions) source term version, while the other one initial condition version.

5.1.1.1 Analytical solution for source term version

Equation (5.3) can also be solved by taking the Fourier transform in space and Laplace transform in time. With all zero initial conditions, after taking the Laplace transform in time, Eq. (5.3) can be written as
\[
\tau_c \tau_R s^2 \hat{e} + \tau_R \hat{e} + \tau_R \mu v \frac{\partial \hat{e}}{\partial x} = \hat{e}^0 - \hat{e} - \tau_c \tau_R \mu v s \frac{\partial \hat{e}}{\partial x} + \tau_R \hat{Q},
\] (5.4)

where \( s \) is the Laplace transform variable. Next, after taking the Fourier transform in space, Eq. (5.4) becomes
\[
\tau_c \tau_R s^2 \hat{\tilde{e}} + \tau_R \hat{\tilde{e}} + i\xi \tau_R \mu v \hat{\tilde{e}} = \hat{\tilde{e}}^0 - \hat{\tilde{e}} - i\xi \tau_c \tau_R \mu v s \hat{\tilde{e}} + \tau_R \hat{\tilde{Q}},
\] (5.5)

where \( \xi \) is the Fourier transform variable. Thus, we can rewrite Eq. (5.5) to obtain the relation between \( \hat{\tilde{e}} \) and \( \hat{\tilde{e}}^0 \):
\[
\hat{\tilde{e}} = \frac{\hat{\tilde{e}}^0 + \tau_R \hat{\tilde{Q}}}{1 + \tau_R s + \tau_R \tau_c s^2 + i\xi \tau_R (1 + \tau_c s) \mu}.
\] (5.6)

In this equation, \( \hat{\tilde{Q}} \) is the source term given by Eq. (4.2). It becomes \( \hat{\tilde{Q}} = 2\pi \delta (\xi - 2\pi/P) \) after the transforms, where \( \delta \) is the Dirac delta function. Plugging this term into Eq. (5.6) and applying the
gray assumption in Eq. (3.18) yield
\[
\hat{\varphi}^0 = \frac{1}{2} \exp(i \lambda x) \int_{-1}^{1} \frac{\hat{\varphi}^0 + 2 \pi \tau R \delta(\xi - 2 \pi/P)}{1 + \tau R s + \tau R \tau_c s^2 + i \lambda \nu \tau R (1 + \tau_c s) \mu} d\mu.
\] (5.7)

This equation can be inversely transformed back into the space domain, so that we have
\[
\hat{\varphi}^0 = \frac{1}{2} \exp(i \lambda x) \int_{-1}^{1} \frac{\hat{\varphi}^0 + \tau R}{1 + \tau R s + \tau R \tau_c s^2 + i \lambda \nu \tau R (1 + \tau_c s) \mu} d\mu,
\] (5.8)

where \( \lambda = 2 \pi/P \) is the wavenumber of the grating pattern. Taking the integral in Eq. (5.8) yields
\[
\hat{\varphi}^0 = \frac{A}{1 - A} \exp(i \lambda x), \quad \text{where} \quad A = \frac{\tan^{-1}\left(\frac{\text{Kn}(1+R_s)}{1+R_s s^2}\right)}{\text{Kn}(1+R_s)},
\] (5.9)

where \( \text{Kn} = 2 \pi \nu \tau R / P \) is the Knudsen number [39], \( R = \tau_c / \tau_R \) is the ratio between \( \tau_c \) and \( \tau_R \). Equation (5.9) can be inverted back to time domain through numerical inverse Laplace transform [50], as will be shown later.

### 5.1.1.2 Analytical solution for initial condition version

For this setup, Eq. (5.3) can also be solved by taking the Fourier transform in space and Laplace transform in time. Compared with the source term version, the nonzero initial conditions introduce extra terms in the solution when applying the Laplace transform. Similarly, we have
\[
\tau_c \tau_R \left( s^2 \hat{\varphi} - s \hat{\varphi}(0, x) \right) + \tau_R \left( s \hat{\varphi} - \hat{\varphi}(0, x) \right) + \tau_R \mu \frac{\partial \hat{\varphi}}{\partial x} = \hat{\varphi}^0 - \hat{\varphi} - \tau_c \tau_R \mu \frac{\partial}{\partial x} \left( s \hat{\varphi} - \hat{\varphi}(0, x) \right),
\] (5.10)

then we apply the Fourier transform in space to obtain
\[
\tau_c \tau_R \left( s^2 \hat{\varphi} - s \hat{\varphi}(0, \xi) \right) + \tau_R \left( s \hat{\varphi} - \hat{\varphi}(0, \xi) \right) + i \xi \tau_R \mu \hat{\varphi} = \hat{\varphi}^0 - \hat{\varphi} - i \xi \tau_c \tau_R \mu \left( s \hat{\varphi} - \hat{\varphi}(0, \xi) \right),
\] (5.11)

With \( \hat{\varphi}(0, \xi) = 2 \pi \delta(\xi - 2 \pi/P) \), we can rewrite Eq. (5.11) as
\[
\hat{\varphi} = \frac{\hat{\varphi}^0 + (\tau_c s + 1 + i \xi \tau_c \mu) 2 \pi \tau R \delta(\xi - 2 \pi/P)}{1 + \tau R s + \tau R \tau c s^2 + i \lambda \nu \tau R (1 + \tau_c s) \mu}.
\] (5.12)

Apply the gray assumption in Eq. (3.18) and take inverse Fourier transform to obtain
\[
\hat{\varphi}^0 = \frac{1}{2} \exp(i \lambda x) \int_{-1}^{1} \frac{\hat{\varphi}^0 + \tau R \tau_c s + \tau R + i \lambda \nu \tau R \tau_c \mu}{1 + \tau R s + \tau R \tau c s^2 + i \lambda \nu \tau R (1 + \tau_c s) \mu} d\mu.
\] (5.13)

Taking the integral in Eq. (5.13) yields
\[
\hat{\varphi}^0 = \frac{(Rs + 1) A + \frac{R}{1+Rs} \left[ 1 - \left( 1 + s + Rs^2 \right) A \right]}{1 - A} \exp(i \lambda x),
\] (5.14)

where \( A \) is still given by Eq. (5.9).
5.1.1.3 Comparison between the two versions

These two cases have been simulated respectively using the gray BTE (simply by setting \( R = 0 \)), and the results are identical due to the explanation given at the beginning of this section. However, when it comes to the EG-BTE, difference appears if the grating period \( P \) decreases to some certain scale (Fig. 5.1).

![Graph showing comparison between solutions calculated with different conditions. The solid lines represent the solutions calculated by applying source term and zero initial conditions (Eq. (5.9)), while the dashed lines stand for the solutions obtained by applying spatially periodic initial conditions (Eq. (5.14)).](image)

Figure 5.1: Comparison between solutions calculated with different conditions. The solid lines represent the solutions calculated by applying source term and zero initial conditions (Eq. (5.9)), while the dashed lines stand for the solutions obtained by applying spatially periodic initial conditions (Eq. (5.14)).

The solid lines in Fig. 5.1 represent the results calculated with source term version solution Eq. (5.9), while the dashed lines show the results calculated by initial condition version solution Eq. (5.14). A clear absorption process can be observed in each of the solid lines as the initial increase of the temperature. The typical time of such process is \( 1 \sim 2 \) ns, differentiating it from the time scale for electro-phonon coupling (within several ps). In large grating period cases, this distinction is insignificant because the two calculated signals still maintain the same decay rate and the experiment time is adequately long to omit this process. But in small grating period cases, \( P = 2 \) \( \mu \)m for example, this process initiates the departure of the signal calculated by the source term version solution from the monotonic decaying signal obtained by the initial condition version solution that more commonly observed and recognized. Physically, the infinitely quick absorption is inappropriate. The obvious heating processes in the experimental data by Johnson et al. also illustrate this point. From this perspective, the source term is a better mathematical description of this experiment.

5.1.1.4 The effective thermal conductivity

Based on the EG-BTE, Wang [9] showed that an ETC is still accessible in the situations with some approximations. Physically, these approximations correspond to the case where the grating
period is not too small (roughly $Kn < 0.5$) and the time of interest is not too short (roughly longer than 10 ns). These two approximations are valid in this study since the experiments by Johnson et al. [36] are simulated, where the maximal $Kn$ is roughly 0.05 and the time for thermal decay signal is more than 25 ns. Under these two assumptions, the ETC given by Wang et al. is offered:

$$
\kappa_{\text{eff}} = \frac{3\kappa}{Kn^2} \frac{1 - \tan^{-1}(Kn)}{1 - R \left[ 1 - 2Kn^2 + \left( Kn^2 - 1 \right) \frac{\tan^{-1}(Kn)}{Kn} \right]}.
$$

(5.15)

Based on Eq. (5.26), the thermal decay rate $\gamma$ can be calculated and plugged into the exponential decay term in the trial solution in 1D TTG part of Chapter 4. We have compared this result with EG-BTE solutions of two versions and confirmed that they are almost the same in the time and length scale of the experiments.

### 5.1.2 Comparison with experiments

For the 1D TTG experiments by Johnson et al. [36] on silicon thin films, as shown in Chapter 4, the EG-BTE produces almost identical results as the TPHC model. This also illustrates the validity of the derivability assumption in Eq. (2.28). Figures 5.2 and 5.3 show the comparisons.

![Figure 5.2: (a). The thermal decay rate and (b). size-dependent effective thermal conductivity of silicon thin films measured from 1D TTG varies with thermal grating periods.](image)

However, as the grating period shrinks to a smaller value, the time scale of the thermal decay curve will also be smaller. As a result, the two assumptions to obtain an ETC do not hold anymore. We proceed to these situations in the next subsection.

### 5.1.3 Extreme 1D TTG cases for the observation of wave-like behavior

Due to the optical limit of heating laser, the extremely small grating pattern is technically difficult to obtain. This fact hinders the progress of study on extreme cases in TTG experiments where the nondiffusive effects are expected to be much stronger. Although there lacks published experimental results regarding the exploration in such situations, Collins et al. [39] applied both...
gray BTE and spectral BTE with phonon dispersion and lifetime data from density functional theory (DFT) calculations, offering some insight of nondiffusive heat transfer in 1D TTG with very small grating periods. They observed oscillatory behavior of the surface temperature and illustrated this behavior with a group of noninteracting phonons traveling inside the gray medium. In addition, they applied a nondimensionalized form of the models so that it could be generally applicable for all materials.

5.1.3.1 Oscillatory temperature by the gray BTE

Based on the analytical solution given in either Eq. (5.9) or Eq. (5.14) with \( R = 0 \) for the gray BTE, the nondimensionalized results are shown in Fig. 5.4.

As comparison, the Fourier’s law solution can be obtained by setting \( \Lambda_B = 0 \) in Eq. (4.5) and plugging this decay rate into the trial solution:

\[
T = T_0 \exp(i \lambda x - \lambda^2 \kappa/c_v t) = T_0 \exp(i x^* - \text{Kn}^2 \tau_R^2/3).
\]  

Equation (5.16) is nondimensionalized with \( x^* = \lambda x \) and \( t^* = t/\tau_R \) to compare to Ref. [39]. The analytical solution by the gray BTE is calculated by the numerical inverse Laplace transform (NILT) MATLAB code. Collins et al. offered two ways to solve the gray BTE, and inverse Fourier transform was applied in one method of the two. We already compared their results and our solution calculated by the NILT code, and found them to be exactly overlapped, as expected. Thus, we validate our results.

The analytical solutions of the gray BTE behaves strongly oscillatory especially when \( \text{Kn}=100 \). In this case, the solution of the gray BTE is approaching the ballistic limit suggested by Collins et al., though the grating period here is nonphysically smaller, i.e., several nanometers. However, this possibly implies the observation of wave-like behavior of phonon transport at room temperature. The oscillatory temperature field serves as a sign of phonon propagation and ballistic heat transfer,
as has been observed at low temperature, which will be mentioned later in this chapter.

5.1.3.2 EG-BTE vs. spectral BTE

Furthermore, Collins et al. also showed the solutions by spectral BTE on 1D TTG using silicon. The application of phonon dispersion improves the prediction on the ETC compared to the gray BTE. Unfortunately, there was no mentioning about the wave-like behavior predicted by the spectral BTE.

Here, we implement the EG-BTE on these artificial cases and compare its results to the spectral BTE results by Collins et al. It should be noted that Collins et al. also presented the results by the two-fluid model, which categorizes phonons into low-frequency and high-frequency groups. This model agrees pretty well with the spectral BTE when $P > 1 \mu m$, and thus its results are not shown in the following comparison to avoid repeating.

Although the source term version solution of the EG-BTE better illustrates the physics, however, since the 1D TTG signals in Ref. [39] all start with 1, initial condition version solutions are plotted in Fig. 5.5 for comparison with other models.

In Fig. 5.5, Fourier’s law dramatically overpredicts the thermal decay, as usual, while the TPHC model and EG-BTE suggest a much slower thermal decay compared to the results from
Figure 5.5: The comparison between the 1D TTG results by different models on silicon thin film [39]. The red dash-dot-dot lines represent Fourier’s law (diffusive limit) in all figures, while the green dash-dot lines show the solutions by the TPHC model. The spectral BTE results by Collins et al. [39] are presented by the blue dashed lines, and the EG-BTE results are given as the solid lines. Four different grating periods are shown: (a) $P = 0.2 \, \mu m$, (b) $P = 1 \, \mu m$, (c) $P = 2 \, \mu m$, and (d) $P = 20 \, \mu m$.

Despite a lack of spectral BTE results on very small grating periods, an oscillatory behavior similar to the solutions by gray BTE in Fig. 5.4 is still desirable. Instead of applying the gray BTE, the solutions by the EG-BTE of two versions (Eqs. (5.9) and (5.14)) are simulated. However, no oscillation has been observed in the results by Eq. (5.14) so that they are not shown here. In
Figure 5.6: EG-BTE result by Eq. 5.9 with grating period \( P = 0.1 \mu m \). High frequency oscillatory behavior of temperature within 0.1 ns marks the wave-like behavior of heat transfer due to phonon transport.

Fig. 5.6, a grating period as small as \( P = 0.1 \mu m \) corresponding to \( Kn=2.63 \) is simulated by Eq. (5.9) and shown. Unlike the results shown in Fig. 5.4 where the oscillation has a large amplitude and low frequency, this result oscillates with relatively small amplitude and much higher frequency. Moreover, this oscillation in the EG-BTE result is damping soon. A better view of oscillation within 0.1 ns is offered in the small window in Fig. 5.6. Consequently, we consider that it is the source term that leads to the oscillatory behavior in the EG-BTE. In other words, the abrupt change of the system state from the equilibrium gives rise to the oscillatory behavior.

As indicated by Fig. 5.6, a very sensitive detector with sufficiently small noise is required if the high frequency and small amplitude oscillation is to be collected. Moreover, it should be noted that similar phenomenon to Fig. 5.1 is also found here: the overall amplitude (maximal temperature rise) is as small as 0.0063 if the overall amplitude for the gray BTE is referred to as 1. Physically, this means the ultrashort stimulated heating may not lead to a detectable response from the material, suggesting the difficulty on top of the expense to reach higher spatial resolution or smaller grating sizes. The detection of wave-like energy transport at room temperature is hard.

5.2 Applications of the EG-BTE on 2D TTG experiments

Similarly, the EG-BTE can be applied on the 2D TTG experiments. Since the physical situation and mathematical description have been given in Chapter 4, we here also directly derive the analytical solutions based on the EG-BTE.

5.2.1 Analytical solution by 2D EG-BTE

In the EG-BTE, the source term can still be written as Eq. (4.7) since the same problem is studied. Starting from Eq. (2.25), the EG-BTE in energy form can be specialized for 2D situations.
Assuming that in Cartesian coordinate, the phonon group velocity can be written as

\[
v = v (\sin \theta \cos \phi \hat{x} + \sin \theta \sin \phi \hat{y} + \cos \theta \hat{z})
\]  

(5.17)

with \(\hat{x}, \hat{y}, \hat{z}\) being the unit directional vector for \(x, y, z\) directions, respectively. Similarly, the gradient term of phonon energy density \(e\) can be expressed as

\[
\nabla e = \frac{\partial e}{\partial x} \hat{x} + \frac{\partial e}{\partial y} \hat{y} + \frac{\partial e}{\partial z} \hat{z}.
\]  

(5.18)

For 2D situations, assuming the variation of \(e\) along \(z\) direction is zero \(\frac{\partial e}{\partial z} = 0\), thus the 2D EG-BTE can be written as

\[
\tau_c \frac{\partial^2 e}{\partial t^2} + \frac{\partial e}{\partial t} + v \left( \sin \theta \cos \phi \frac{\partial e}{\partial x} + \sin \sin \phi \frac{\partial e}{\partial y} \right) = -e - e^0 - \tau_v \frac{\partial}{\partial t} \left( \sin \theta \cos \phi \frac{\partial e}{\partial x} + \sin \sin \phi \frac{\partial e}{\partial y} \right) + Q.
\]  

(5.19)

Similar to what has been done in the 1D TTG solution, we here still take the Fourier transform in space and Laplace transform in time to obtain solution of \(e\) in the frequency domain. The derivation is very similar to 1D TTG part, so we skip it and directly give the result:

\[
\tilde{e} = \frac{\tilde{e}^0 + \tau_R \tilde{Q}}{1 + \tau_R \tau_c \xi^2 + i \nu \tau_R \sin \theta (1 + \tau_c) \left( \xi_x \cos \phi + \xi_y \sin \phi \right)}.
\]  

(5.20)

With the gray-medium assumption given in Eq. (3.18), we have

\[
\tilde{e}^0 = \frac{1}{4\pi} \int_0^{2\pi} \int_0^\pi \left( \tilde{e}^0 + \tau_R \tilde{Q} \right) \sin \theta d\theta
\]  

\[
\]  

(5.21)

After integrating this equation, the \(\tilde{e}^0\) can be solved as

\[
\tilde{e}^0 = \frac{\tau_R \tilde{Q} \tilde{A}(s, \xi)}{1 - A(s, \xi)}, \quad \text{where} \quad A = \frac{\tan^{-1} \left( \frac{1 + \tau_c s}{1 + \tau_c \nu \tau_R} \nu \tau_R \xi_y \right)}{(1 + \tau_c) \nu \tau_R \xi_y}.
\]  

(5.22)

One may notice that the solution given in Eq. (5.22) is still in the same form of the 1D solution except that \(\xi = \sqrt{\xi_x^2 + \xi_y^2}\). However, this \(\tilde{e}^0\) also contains the spatial frequency in \(y\) direction that will be integrated to obtain the signal given in Eq. (4.16) with \(\Delta \tilde{\beta} = \tilde{e}^0\). On the other hand, \(\tilde{Q} = 4\pi Q_0 \delta (\xi_x - \lambda) \frac{\beta_1}{\beta_1^2 + \xi_y^2}\). Thus, the signal can be written as

\[
\tilde{H}(s, \xi_x) = \int_0^\infty A(s, \xi) \frac{\tilde{Q}_R}{1 - A(s, \xi)} \frac{2\beta_2}{\beta_1^2 + \xi_y^2} d\xi_y
\]  

\[
= 8\pi Q_0 \tau_R \delta (\xi_x - \lambda) \int_0^\infty A(s, \xi) \frac{\beta_1 \beta_2}{\beta_1^2 + \xi_y^2} d\xi_y.
\]  

(5.23)
Accordingly, the signal can be calculated as

\[
\hat{H}(s, x) = A' \exp(i\lambda x) \int_0^\infty \frac{A(s, \xi)}{1 - A(s, \xi) \left(\beta_1^2 + \xi_y^2\right) \left(\beta_2^2 + \xi_y^2\right)} d\xi_y, \quad (5.24)
\]

where \(A'\) is the amplitude that will be normalized before the comparison, and \(\xi = \sqrt{\lambda^2 + \xi_y^2}\).

For simplification, similar assumptions as applied in the solution by the TPHC model are also applied here to write the signal in Eq. (5.24) into an exponential form:

\[
H(t, x) = A' \exp(i\lambda x) \int_0^\infty \exp\left[-\kappa(\xi) \xi_y^2 t/\tau_R\right] \frac{1}{\left(\beta_1^2 + \xi_y^2\right) \left(\beta_2^2 + \xi_y^2\right)} d\xi_y, \quad (5.25)
\]

where the ETC \(\kappa(\xi)\) contains the integral variable \(\xi_y\). Taking \(\text{Kn} = v\tau_R\xi\) and \(R = \tau_e/\tau_R\), we have ETC similar to the 1D form

\[
\kappa(\xi) = \frac{3\kappa}{\text{Kn}^2} \left(1 - \frac{\tan^{-1}(\text{Kn})}{\text{Kn}}\right) \frac{1}{1 - \lambda/R \left[1 - 2\text{Kn}^2 + \left(\text{Kn}^2 - 1\right) \tan^{-1}(\text{Kn})\right]}.
\]  

(5.26)

Carrying out the quadrature in Eq. (5.24) with Eq. (5.26) yields the final solution of the optical signal. It should be noted that due to the integration of \(\xi_y\), there is no explicit expression of ETC in time and space domain.

However, there is still an absorption process similar to the phenomena observed in 1D TTG solved by applying the source term in EG-BTE. An alternative solution with initial conditions specified as \(e(t = 0, x) = e_0 \exp(i\lambda x - \beta_1 y)\) and \(\partial_e/\partial t\big|_{t=0} = 0\). The solution of \(e\) can be obtained following the process given in the 1D TTG part. We skip the derivation and offer the solution here

\[
\tilde{e} = \tilde{e}_0 + \frac{1 + \tau_e s + iv\tau_e \sin \theta (\xi_x \cos \phi + \xi_y \sin \phi)}{1 + \tau_R s + \tau_e s \sin \theta (1 + \tau_c s) (\xi_x \cos \phi + \xi_y \sin \phi)} \tau_R \tilde{e}(0, \xi_x, \xi_y).
\]

(5.27)

Similar to Eq. (5.21), \(\tilde{e}_0\) can be solved

\[
\tilde{e}_0 = \frac{\tau_e \left[1 + \tau_c s + (1 + \tau_e s) \tau_R A - \frac{\tau_e \left[1 + \tau_R s + \tau_e \tau_R s^2\right]}{1 + \tau_e s} A\right]}{1 - A} \tilde{e}(0, \xi).
\]

(5.28)

Accordingly, the signal can be calculated as

\[
H(t, x) = A' \exp(i\lambda x) \int_0^\infty \frac{\tau_e \left[1 + \tau_c s + (1 + \tau_e s) \tau_R A - \frac{\tau_e \left[1 + \tau_R s + \tau_e \tau_R s^2\right]}{1 + \tau_e s} A\right]}{1 - A \left(\beta_1^2 + \xi_y^2\right) \left(\beta_2^2 + \xi_y^2\right)} d\xi_y.
\]

(5.29)

Eq. (5.29) can be inversely transformed back into the time domain using the NILT code, and numerically integrated to yield the final result for the signal. We will compare this result with experiments and the spectral BTE later.
Figure 5.7: Comparison between analytical solution by the EG-BTE and experimental results: (a) by Johnson et al. [40] with the thermal grating periods are $P = 10 \mu m, 4.9 \mu m, 3.2 \mu m, 2.05 \mu m$ from high to low, and (b) by Johnson et al. [37] with the thermal grating periods are $P = 4.9 \mu m, 3.7 \mu m, 2.8 \mu m, 2.4 \mu m$ from high to low.

### 5.3 Validations and predictions for 2D TTG

Similar to the validations in Chapter 4, we also validate the EG-BTE with both experimental and theoretical results. In the scale of the validation, the EG-BTE again performs very similar to the TPHC model, similar to the way in the 1D TTG cases. For this reason, we directly show the comparison here.

#### 5.3.1 Comparison with experiments

Figure 5.7 shows the comparison between the signal simulated by Eq. (5.24) and experiments. The EG-BTE provides very similar results to the TPHC model, as expected. All the thermophysical properties are given in Table 5.1.

<table>
<thead>
<tr>
<th>Material</th>
<th>$c_v$ ($J/m^3K$)</th>
<th>$v$ (m/s)</th>
<th>$\kappa_{bulk}$ (W/mK)</th>
<th>$\tau_e$ (ns)</th>
<th>$\beta_1$ (nm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicon</td>
<td>$1.66 \times 10^6$</td>
<td>6400</td>
<td>148</td>
<td>0.45</td>
<td>1/580</td>
</tr>
<tr>
<td>GaAs</td>
<td>$1.75 \times 10^6$</td>
<td>3700</td>
<td>45</td>
<td>3.36</td>
<td>1/150</td>
</tr>
</tbody>
</table>

Table 5.1: Thermophysical properties of materials involved in 2D TTG study ($\beta_2 = \beta_1$) for the EG-BTE.

#### 5.3.2 Comparison with spectral BTE

In addition to the experimental results, we also compare the EG-BTE and the spectral BTE by Minnich [41]. But the artificial cases as shown in Chapter 4 are not shown here, since oscillatory behavior and the absorption process are already observed for those grating periods.
Figure 5.8: Comparison between results from 2D EG-BTE (dashed lines) and synthesized temperature decay curve by Minnich [41] based on spectral BTE with phonon dispersion calculated by first principle (solid lines).

5.4 Some discussions on extreme cases

According to the discussion on \( \zeta_y \) in Chapter 4, when the penetration depth is very large, nondiffusive heat transfer in the in-plane direction dominates the reduction of ETC so that 1D approximation can be made. This fact is also pointed out by Minnich [41], who stated that transport in the cross-plane direction can be neglected when the penetration depth is at least three times greater than the grating period. By assuming this, the 2D TTG can be reduced to 1D TTG so that the extreme cases features perform similarly to what have already been shown in the 1D TTG section of this chapter. The oscillatory temperature and the difference between source term version and initial condition version of the EG-BTE are also be found.

Besides an extremely large penetration depth, the other end of the limit is an extremely small penetration depth. In this situation, the nondiffusive behavior is strongly contributed by the transport in the cross-plane direction. A highly dynamic heating process can be captured by calculating the signal using Eq. (5.25), while Eq. (5.29) describes a signal that remains still at initial moment \( t = 0 \). Physically, Eq. (5.25) better describes the heating process and is preferred to be applied here. However, no oscillatory signal has been observed if the grating period is large enough. Instead, both the amplitude of the signal and the absorption time to reach maximal signal amplitude vary with different penetration depths \( \beta_1^{-1} \) even if \( P \) is fixed, suggesting the possibility to decouple the effects. The increase of \( \beta_1^{-1} \) gives rise to the increase in both the amplitude and the absorption time to reach the peak. The increase of amplitude performs high sensitivity to \( \beta_1^{-1} \), while the absorption time is only slightly influenced by \( \beta_1^{-1} \). This increase of amplitude can be mathematically seen by calculating the total energy deposited by the laser at each \( x \) location as:

\[
Q_{tot}(t, x) = Q_0 \exp(i\lambda x)\delta(t) \int_0^\infty \exp(-\beta_1 y)dy = \frac{Q_0}{\beta_1} \exp(i\lambda x)\delta(t),
\]  

\[(5.30)\]
which is proportional to $\beta^{-1}$. On the other hand, the delay of the absorption time could be qualitatively explained by the heat flux reduction in the cross-plane direction due to the nondiffusive effects. For a fixed thermal grating period, the energy deposited by the laser distributes more evenly with a larger $\beta^{-1}$, leading to a weaker nondiffusive transport, or suppression of energy transport, in the cross-plane direction. As a result, a more efficient transport spreads out the energy absorbed by the material near the surface more quickly, resulting in a slower local accumulation of energy. Since the variation of the amplitude of the signal goes across several magnitudes, hindering the visualization, the results for different penetration depths are not shown here.

5.5 Applications of the EG-BTE on the MG-X-ray experiments

The experimental setup and physical situation of the MG-X-ray experiments have been described in Chapter 4, so we skip it here. In addition, the metal heater subdomain given in Fig. 4.7(b) is still modeled by Fourier’s law due to the same reason specified in Chapter 4, but the substrate subdomain will be modeled using the EG-BTE. Consequently, we only give the boundary conditions and initial conditions for the substrate subdomain here.

Similar to the boundary conditions and initial conditions for the TPHC model, the boundary condition at the interface for the heater subdomain can still be given as

$$-\kappa_h \frac{\partial T}{\partial n} = \begin{cases} T_h - T_s, & \text{on heater-substrate interface,} \\ 0, & \text{else,} \end{cases}$$

(5.31)

where $T_s$ is the temperature of the subdomain at corresponding position. This temperature can be related to the local equilibrium phonon energy density $e^0$ through Eq. (2.26).

For the substrate subdomain, there is still insulation at $y = y_{\text{max}} \to \infty$ due to the thickness of the substrate. At $y = 0$, the boundary condition is similar to Eq. (5.31):

$$q_y = \begin{cases} T_h - T_s, & \text{on heater-substrate interface,} \\ 0, & \text{else.} \end{cases}$$

(5.32)

It should be noted that the EG-BTE does not require additional boundary condition as the TPHC model. Thus there is no boundary condition at $y = 0$ for $q_x$. At left and right boundaries shown in Fig. 4.7(b), periodic boundary conditions are implemented.

Moreover, there is no numerical issue due to the two singularity points located at the two corners of the heater-substrate interface (see Fig. 4.7(b)). Hence, the heat flux is rigorously given by Eq. (5.32) without smoothed.

As the initial conditions, the normalized temperatures in the nickel heater and the substrate are specified as $T_h = 1$, $T_s = 0$ and $\partial T_s/\partial t = 0$, respectively.

With the auxiliary conditions specified, the EG-BTE is numerically solved by LBM. The implementation of boundary conditions has been introduced in Chapter 3 so that it is not skipped here.

5.5.1 Reinterpretation of MG-X-ray experiments on silicon

In Chapter 4 we showed the comparison between the results by the TPHC model and the experimental data by Siemens et al. [42] on sapphire substrate. The EG-BTE again offers similar
results so they are not shown here. However, In addition to the experiments on a sapphire substrate, Hoogeboom-Pot et al. [48] also offered MG-X-ray results on a silicon substrate. These MG-X-ray cases are using smaller heater widths down to \( L = 20 \text{ nm} \) with thickness \( \sim 13.5 \text{ nm} \). According to the calculation of \( \zeta = 610.23 \gg 1 \) based on fitted \( \Lambda_B = 7.1 \text{ \mu m} \) for \( L = 20 \text{ nm} \) and \( P = 4L \), the nondiffusive heat transfer is so strong that the assumptions used to derive the TPHC model from EG-BTE already break down. Actually, we already observed abnormal oscillatory temperature around the two corners of the heater using the TPHC model, even if we smoothed out the discontinuity on the interface as in sapphire cases. For silicon, this abnormal spike obscures or even destroys the calculation accuracy of the diffracted signal. Consequently, the EG-BTE is applied for these extremely small cases for the study on nondiffusive heat transfer and extraction of interface resistivity.

![Graphs](image)

Figure 5.9: Comparison of the fitting results of the EG-BTE and the experimental data (In Ref. [48] and unpublished) on a silicon substrate. The unit of the thermal interface resistivity \( r \) is \( 10^{-9} \text{ Km}^2/\text{W} \).

Overall, there are three silicon cases published with the nickel heater widths \( L = 60, 30 \) and 20 nm, respectively. Moreover, some additional cases with \( P = 400 \text{ nm} \) rather than the typical fixed \( P = 4L \) period are also provided for \( L = 30 \) and 20 nm. In these cases, the size of the interface now is fixed with the grating periods varying, which is helpful to prove or disprove our hypothesis that the size-dependence of the interface should be defined by their own parameters (i.e., the surface roughness, size, difference of materials) instead of the spacing of the neighboring heaters. Different
The spacings of neighboring heaters do contribute to the nondiffusive heat transfer inside the substrate, and such a contribution is able to be captured by EG-BTE, as shown in the 2D TTG case. The spacing between two neighboring heaters (period) does not affect this property of the interface.

Here the same fitting process as described in the TPHC part is performed to extract the value of $r$ from experimental data. The experimental data include both published data with $L = 60, 30$ and $20$ nm and some unpublished ones (all others). These experimental data still show clear acoustic oscillation due to Rayleigh surface wave, but the noise level is so high that it becomes comparable to the laser-induced acoustic oscillation, especially towards the tail part of the signal. Fortunately, what we are interested in is the thermal decay of the diffracted signal. For this reason, only the thermal decay component of the signal is plotted to compare with the experimental measurements in Fig. 5.9 and Fig. 5.10, assuming the noise is random and has a zero average.

For the cases with fixed spacing-to-linewidth ratio $P = 4L$, the best-fitted interface resistivity $r$ increases monotonically with the shrinking $L$. This monotonic behavior differs from the unexpected trend observed in the sapphire grating cases. The fitting results by Fourier’s law, on the other hand, are provided by Hoogeboom-Pot et al. Among these results by Fourier’s law, a kink peak appears around $L = 190$ nm, indicating that $r$ fitted by Fourier’s law should decrease as $L$ shrinks. This interesting and implicative mismatch is left for further study.

For the cases with a fixed linewidth $L$ and different grating period $P$, numerical results by EG-BTE also fit the experimental data in Fig. 5.10. Here, the comparison and the size-dependence of the fitted $r$ inspire us to propose the hypothesis mentioned at the beginning of this section: the effective interface resistivity $r$ in the MG-X-ray experiment depends only on the size of the interface (heater width $L$). Specifically, the fitted $r = 3.5 \times 10^{-9}$ Km$^2$/W in both cases with $L = 20$ nm and different $P$. Close values are obtained in the fitting with larger $P$, including the case with $L = 30$ nm and $P = 120$ nm. However, a mismatch is shown in Fig. 5.10: three out of the four cases can be fitted with $r = 3.5 \times 10^{-9}$ Km$^2$/W by the EG-BTE, while the case where $L = 30$ nm and $P = 400$ nm cannot. In this figure, the green dashed-dot line represents the simulated signal with $r = 7.5 \times 10^{-9}$ Km$^2$/W, and the red dashed line stands for the simulated signal with $r = 3.5 \times 10^{-9}$ Km$^2$/W. Although such a mismatch may disprove our hypothesis, it could also be attributed to the experiment uncertainty, especially considering that only one set of experimental data is given in the paper.

<table>
<thead>
<tr>
<th>$L$</th>
<th>250 nm</th>
<th>100 nm</th>
<th>60 nm</th>
<th>40 nm</th>
<th>30 nm</th>
<th>20 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_{EG-BTE}$</td>
<td>2.8</td>
<td>2.8</td>
<td>3.0</td>
<td>3.3</td>
<td>3.5</td>
<td>3.5</td>
</tr>
</tbody>
</table>

Table 5.2: Summary of fitted interface resistivity $r$ with different nickel heater width $L$. The unit of $r$ is $10^{-9}$ Km$^2$/W.

5.5.2 Discussion

There are two significant advantages for MG-X-ray experiment: 1). the size of interface is controllable so that the systematic study on nondiffusive heat transfer across the interface can be performed; 2). the application of nano-sized strips heater extends the limit of the grating sizes to tens of nanometers, facilitating the study on strong nondiffusive heat transfer inside the substrate. We here report the observation of some interesting but not yet validated phenomena.
5.5.2.1 Size-dependence of the interface resistivity

Effective interface resistivity $r$ fitted by both the TPHC model and EG-BTE shows size-dependent behavior, indicating that the heat transport across the interface may also be nondiffusive. In sapphire case for example, when the heater width in the MG-X-ray experiments is larger than 810 nm ($P = 4L > 3.2 \mu m$), the value of $\zeta$ is less than 0.01. The effects from the nondiffusive heat transfer both across the interface and inside the substrate are negligible. Thus heat transfer is diffusive. For diffusive heat transfer, there is a constant thermal interface resistivity. As heater size $L$ decreases, there is monotonically increasing nondiffusive effects according to 2D TTG study, while the effective interface resistivity unexpectedly decreases. The cause should most possibly be attributed to the size-dependent nondiffusive heat transfer across the interface, if not from inside of the substrate. To explain this phenomenon, the heat transfer mechanisms across the interface should be studied.

At the heater-substrate interface, there are different scattering mechanisms for diffusive and
nondiffusive heat transfer, which can be described by diffuse mismatch model (DMM) and acoustic mismatch model (AMM) [51], respectively. Therefore, for different contributions of nondiffusive heat transfer, there are different values of the thermal interface resistivity. For the same interface, there is higher thermal interface resistivity (lower conductance) in nondiffusive heat transfer than diffusive heat transfer, as reported in the previous study of nondiffusive heat transfer in TDTR experiments [52]. This can explain the monotonic increase of the TPHC results on the thermal interface resistivity as \(L\) decreases from 810 nm to 190 nm as shown in Fig. 4.11. However, as \(L\) decreases further from 190 nm to 80 nm, there is significant decrease in the TPHC results of the thermal interface resistivity. The similar results are also reported by Hoogeboom-Pot et al. in their MG-X-ray experiments using Fourier’s law for data analysis [48]. The abnormal decrease in the thermal interface resistivity for smaller heater size is called collective diffusive effects by Hoogeboom-Pot et al.. Recall that we observe monotonically increasing gap between Fourier’s law and the TPHC results in 2D TTG, possibly implying that such an unexpected phenomenon might be due to the interface. In fact, at the thermal interface, a “radiative” boundary condition defined by the resistivity across the interface is used in this study. However, this boundary condition is developed under the assumption of diffusive heat transfer, and unable to capture the physics of nondiffusive phonon scattering and transmission at the interface. In MG-X-ray experiments, there is mixed diffusive and nondiffusive heat transfer for the cases where \(L < 810\) nm, if the phonons can be simply categorized into two groups. The contributions from diffusive phonons and nondiffusive phonons are unknown and dynamic in time, which makes it challenging to propose an accurate thermal boundary condition to account for both diffusive and nondiffusive heat transfer.

Generally, the interface resistivity is different at different locations of the interface. Around corner of the heater where the symmetry is broken, for example, a higher resistivity may appear since it causes the mismatch of the lattice structure. In the interface of small sizes, the influence regime of higher resistivity weighs more in the overall resistivity so that the resistivity fitted is of a larger value. On the other hand, if the size of the interface is large enough that the influence regime
of the corner contributes negligibly to the whole regime, the resistivity converges to a constant value known as diffusive value. Of course, other factors such as the surface roughness cannot be ruled out.

In conclusion, the cause and mechanism of size-dependence of interface resistivity still require further studies that will be our future work.

5.5.3 Thermal wave inside the substrate

By definition, phonon describes lattice vibration modes of elastic solid crystals in quantum mechanical view and travels as thermal wave. This wave behavior distinguishes the energy transport from mass transport where the particles travel ballistically. The observation of the wave behavior is expected as it naturally describes the lattice vibrations, and may directly unify the quantum mechanical view of heat transfer and the traditional heat conduction. In the 1D TTG section of this chapter, we discover the wave-like behavior of temperature through analytical solution of both gray BTE and EG-BTE, but the reconstruction of heat flux field is hard to obtain analytically, if not impossible. Here, with the phonon energy solved by LBM, the heat flux field is accessible through Eq. (2.27). The cross-plane heat flux ($q_y$) profiles along the centerline in Fig. 4.7(b) for two cases where $L = 20$ nm at $t = 1$ ns are plotted in Fig. 5.12. The two profiles are normalized by the maximal value of $q_y$ in $P = 80$ nm profile.

![Figure 5.12: Comparison of size-dependent thermal interface resistivity changing with nickel heater width $L$ between TPHC model and Fourier’s law.](image)

The oscillatory cross-plane heat flux exists for both cases. This oscillatory feature decays in amplitude towards the depth into the substrate, and is influenced by the spacing $P$ of the neighboring heaters. A very large spacing $P = 400$ nm suggests treating the heater as isolated heat source [48] since the heater width $L = 20$ nm is fixed in these two cases, while the spacing $P = 4L = 80$ nm still suggests the periodicity is important. In Fig. 5.12, the amplitude of $q_y$ in the isolated heater case decays faster than in the periodic case. In addition, the maximal amplitude in the periodic case appears at certain depth rather than the vicinity of the surface. Both observed features indicate that
the oscillation is enhanced by the neighboring heater and offer tips for the direct observation of such a wave-like behavior: spacing is one important factor in experiment with similar setups.

This observation is very interesting and unconventional, thoroughly calling the definition of thermal conductivity into question in this time and length scale. If proved true, it means that the EG-BTE works in an amazingly wide length and time scale covering both ballistic phonon transport and diffusive heat transfer. Unfortunately, to experimentally capture this wave-like behavior requires extremely high sensitivity and resolution of the experimental instrument that may not be available at present. However, similar observation has been reported around half a century ago. During that time, people carried out experiment to measure the thermal wave under low temperature (tens of kelvin or lower). In next section, the EG-BTE will be applied to study the thermal wave under low temperature.

5.6 Applications of the EG-BTE on thermal wave experiments

In this section, the EG-BTE is applied to explain the observation of thermal wave at low temperature, where the thermal transport behaves ballistically and the phonon transport is experimentally captured as the temperature wave. The thermal wave is successfully simulated by the EG-BTE using phonon discrete ordinate methods developed in Chapter 3, and qualitatively compared to the experimental observation. The agreement between the EG-BTE simulation results and experimental observation marks EG-BTE’s ability to qualitatively describe the physics of thermal wave, and suggests a similar prediction at room temperature. Through EG-BTE, the seemingly distinct thermal transport mechanisms are unified, providing new understanding of heat transfer.

5.6.1 Introduction

As described in Chapter 1, the essence of phonons is the lattice vibration, which suggests the wave nature of energy transport by phonons. This wave-like behavior is one of the core distinction of energy transport compared to mass transport. Physically, in mass transport, two particles collide with each other when they meet, while two waves by nature pass each other when they come across. The wave-like behavior of energy transport, unfortunately, has not been discovered at room temperature, though there exist results observed deviating from diffusive transport. For example, in this thesis, all the experimental results shown so far either indicate a different ETC from the prediction by Fourier’s law, or demonstrate a slower thermal decay rate compared to Fourier’s law. These nondiffusive observations are truly different from the diffusive heat transfer, but they cannot be classified as the ballistic heat transfer. Therefore, a direct observation of the wave-like behavior for energy transport can bridge the gap between quantum mechanical perspective in nanoscale and the experimental observation at room temperature, and thus consolidate our understanding of thermal transport.

The direct observation of thermal wave, or the evidence of wave-like behavior, has actually been discovered since the middle of last century. By applying a pulsed Joule heating on one side of a very pure bulk dielectric sample at low temperature (~10⁸K), researchers were able to capture a temperature pulse from the other side of the sample [5, 53–55]. The schematic of the experiment is exhibited in Fig. 5.13. In this situation, the temperature pulses propagate ballistically as longitudinal and transverse waves (first sound) with respective speeds. Accordingly, two pulses corresponding to these two polarization modes can be detected in certain temperature range. In addition to the first

80
sound, there exists the second sound [5]. It is a collective temperature pulse with a characteristic velocity determined by the interaction of all the phonon characteristics in the crystal. It becomes detectable when the momentum-conserving phonon collisions (N process) are dominant over the momentum-destroying collisions (U process). In an appropriate temperature range depending on the purity of the sample, this second sound can be experimentally detected as the third peak after two peaks of the first sound in the temperature signal.

In addition to the experimental study, theoretical models including C-V model [11, 12], G-K model [14, 56], the 9 moment theory [57], and hybrid phonon gas model [58], were also developed. However, these models for explaining the thermal wave phenomena are usually not able to give a satisfactory description for other nondiffusive heat transfer experiments. Being able to describe multiple nondiffusive heat transfer experiments like TTG, TDTR [9] and MG-X-ray, EG-BTE is also able to reproduce the characteristics of phonon transport at low temperature, as will be shown in this section.

5.6.2 Validation of phonon $S_N$ method

The 1D EG-BTE, given by Eq. (5.3), has to be solved numerically for this setup. We first validate the the phonon $S_N$ method proposed in this thesis, while leave the boundary and initial conditions for later explanation. The results by LBM are compared with the analytical solution of the gray BTE on 1D TTG, which can be obtained by setting $R = 0$ in Eq. (5.9), to emphasize the discrepancy. In Fig. 5.14, the solid lines represent the the gray BTE analytical solutions by Eq. (5.9) with $R = 0$, while the dashed lines are LBM results. Certain discrepancy can be observed as $Kn$ increases from 0.1 to 0.5. In the MG-X-ray case, Eq. (3.12) still works acceptably if Knudsen number $Kn = \frac{\nu \tau}{P/2} \approx 1$ for the silicon gratings with $P = 80$ nm, considering half period as the MFP for the phonons emitted through the interface. This value may be smaller due to the infinite depth in the cross-plane direction. But as $Kn$ grows increasingly larger to more than 1, LBM fails to offer valid solutions.

On the other hand, phonon $S_N$ method is implemented to solve the gray BTE and compared to the analytical solutions in Fig. 5.15. Phonon $S_N$ method here provides identical solutions as the analytical method through the NILT. Also, the analytical solutions by EG-BTE on 1D TTG with
Figure 5.14: LBM results (dashed lines) gradually depart from analytical solution of 1D TTG (solid lines) as Kn increases. For large Kn number cases, LBM fails to give reliable solutions.

very small grating periods can also be reproduced by phonon $S_N$ method but not shown here due to brevity concerns.

In addition to the governing equation itself, appropriate boundary conditions have to be specified so that the physical situation can be successfully simulated. Ma [58] indicated that the boundary condition for the heating side of the sample can be approximated as a pulsed temperature with certain duration time $\Delta t$. Dreyer et al. [57], on the other hand, suggested a square temperature wave input with two slope transitional edges. Both of the two options suggest a Dirichlet boundary condition with a time-dependent temperature, for which Eq. (3.32) is applicable. The other side of the sample where thermometer is evaporated can be approximately treated as insulated and Eq. (3.35) with $q = 0$. However, certain amount of energy loss should be taken into account when the measurement is taken for a relatively long time (i.e., tens of $\mu$s). Eq. (3.38) is a better choice for such scenario because the energy loss usually depends on the temperature difference and the interface thermal resistance.

In next section, phonon $S_N$ method is applied to solve the thermal wave problem.

5.6.3 Different regimes of thermal wave propagation

According to the experimental studies, the strong ballistic transport can be detected at temperature typically lower than 10$^\circ$K, while the diffusive transport dominates the heat transfer at higher temperature close to 20$^\circ$K. Between these two regimes, transitional regime is contributed by both of the two transport mechanisms. A second sound, falling within the transitional range, is a consequence of the momentum-conserved scattering. It consists of the phonons experiencing N processes with momentum conserved but arriving time delayed. The extra delayed time compared to pure ballistic phonons varies and depends on the details of the scattering processes. Thus, these phonons give rise to the second peak following the pulse representing the first sound of the corresponding polarization. Outside the appropriate regime, these phonons either propagate ballistically to the
Figure 5.15: Solutions of gray BTE at different Kn numbers (a) Kn = 0.25 – 5; (b) Kn = 10; (c) Kn = 100. The solid lines represent the analytical solutions of Eq. (5.14) with $\tau_c = 0$ obtained through NILT, and the square symbols stand for the results by phonon $S_N$ method. The two curves exactly overlap with each other.
other side of the sample, or strongly scattered so that they lose the collective effects and perform diffusively.

Therefore, three different regimes of thermal wave propagation can be divided: (1) ballistic regime (2) transitional regime and (3) diffusive regime, depending on the parameters $\tau_R$ and $\tau_c$. A parametric study can systematically assist the understanding of the division of different regimes, and the physical interpretations of the parameters. Considering that the total phonon energy is a superposition of longitudinal and transverse polarizations, only the transverse component is studied as an example, since the longitudinal component follows a similar way. It is important to note that there does exist the energy exchange between the polarization modes through scattering, for example, a longitudinal acoustic phonon is split into two transverse acoustic phonons [59, 60]. However, this effect is assumed to be weak and omitted in this study.

Here, we claim that the following studies are based on NaF as the sample material of various sizes, and at different environmental temperatures. The sound speed for longitudinal wave is $c_l = 6051$ m/s and transverse wave is $c_t = 3186$ m/s according to Ref. [53, 54].

### 5.6.4 Parametric study

![Image](image.png)

**Figure 5.16:** Thermal wave simulated by the gray BTE using phonon $S_N$ method with different relaxation time $\tau_R$. The size of the sample is 7.9 mm here. The number on each curve marks the corresponding value of $\tau_R$ in $\mu$s for the curve. From the diffusive response to a single ballistic peak signal, the gray BTE predicts no extra peak representing the second sound.

The gray BTE is the fundamental of the EG-BTE since it is a special case of the EG-BTE. The gray BTE is only equipped with one parameter $\tau_R$, simplifying the parametric study. Throughout the possible range of $\tau_R$, the gray BTE results are given in Fig. 5.16. All the simulated signals are normalized by its maximum if not particularly specified, since we are mainly interested in the waveform. The amplitude is sensitive to numerous factors such as input power and temperature [5], which is not the focus of this study. From the diffusive regime with small $\tau_R$ to the ballistic regime with large $\tau_R$, a smooth transition takes place without emerged second peak representing second
Figure 5.17: The EG-BTE results of the transverse component of thermal wave with an intermediate relaxation time $\tau_R = 1.0 \, \mu s$. (a) Emerging of second peak; (b) merging of second peak.
sound. Despite of this, gray BTE does correctly describe two limits: pure ballistic with $\tau_R \to \infty$, and pure diffusive with $\tau_R \to 0$. It may be due to the failure to capture the second sound that the gray BTE cannot describe the transitional scale where ballistic and diffusive mechanisms are competing. For example, we can look back at 1D TTG ETC predicted by the gray BTE in Fig. 5.3. It severely overpredicts the increase of the ETC with increasing thermal grating periods.

However, the EG-BTE results do indicate the discovery of the second sound, as can be seen in Fig. 5.17(a). A second peak, with $\tau_R = 1.0 \mu s$ and $R = 0.04$, is captured and displayed as the blue solid line. This peak arrives much earlier than the diffusive tail, which typically takes more than $5 \mu s$ to arise with this $\tau_R$ value, and later than the first sound peak. Its arriving time also varies with $\tau_c$ rather than fixed as that of the first sound. Both of these characteristics match the previous statements from experimental studies regarding the second sound. Similar to the fact that a suitable temperature range is necessary for the detection of the second sound, we have found that an appropriate $\tau_c$ range is necessary to allow the emerging and merging of the second sound. Small $\tau_c$ leads to the reduction of the EG-BTE back to gray BTE, which does not predict the existence of second sound; while large $R$ redistributes the majority of energy carried by the second sound into the first sound peak as shown by the orange line in Fig. 5.17(a). Thus, the ballistic transport weighs more due to the enhancement by terms with $\tau_c$, and this is the physical interpretation of terms with $\tau_c$.

Figure 5.18: Simulation of the EG-BTE for (a) $\tau_R = 1.0 \mu s$ and (b) $\tau_R = 0.5 \mu s$ with different ratio $R$. The amplitude is not normalized so that the energy redistribution can be visualized.

In detail, the modification on the waveform due to the second order derivative terms is visualized in Fig. 5.17, but Fig. 5.18 more directly illustrates the redistribution of phonon energy by these terms. Since the energy input is strictly the same for all the cases shown in Fig. 5.18, total energy deposited into the computational domain is identical. The change of amplitude is solely due to different contribution from these terms characterized by different $R$, and it is this redistribution that
Figure 5.19: Waveforms with same second-to-first peak amplitude ratio but different $\tau_R$. With a fixed amplitude ratio, the arriving time of the second sound peak varies monotonically, allowing a unique fitting of both $\tau_R$ and $\tau_c$ simultaneously. Larger $\tau_R$ than 1.5 $\mu$s results in no maximum after the first peak, indicating that the second sound peak has been mainly redistributed into the first sound peak.

introduces the etch of the signal after the first sound peak and gives rise to the second sound peak in the EG-BTE. For example, in Fig. 5.18(a), this redistribution effect introduces a better efficiency of the energy transport, since more phonons reach the thermometer side at the first sound speed. In Fig. 5.18(b), the redistribution of phonon energy causes reappearance of the first sound peak that used to be buried by the diffusive tail. Although the first sound amplitude is markedly raised by the effect from these terms, the energy loss due to this redistribution process has negligible impact on the dominant diffusive tail. Therefore, the EG-BTE shares the same limits (ballistic and diffusive) as the gray BTE, but overperforms the gray BTE in transitional regime of thermal wave transport.

Another concern is the feasibility to fit both $\tau_R$ and $\tau_c$ simultaneously using only one experimental trace, if a second sound appears. According to Fig. 5.19, this is feasible with two features in the trace extract: (1) the ratio between the amplitude of the second sound to the amplitude of the first sound and (2) the arriving time of the second sound. The results exhibited in Fig. 5.19 indicate that the arriving time of the second sound varies with $\tau_R$ monotonically once a second-to-first amplitude ratio (i.e., 0.8 here) is specified. The disappearance of the second sound peak unfortunately lowers the accuracy of the fitting, since in those regimes the waveform is insensitive to $R$, or $\tau_c$ in other words.

Outside the window for the second sound, the sensitivity of the results to $\tau_c$ decreases. Around the ballistic limit where $\tau_R$ is large, the change of $\tau_c$ does not introduce significant changes in the waveform compared to the gray BTE results, as can be seen in Fig. 5.20. This is because the heat transfer is approaching the limit of ballistic transport even without the enhancement brought by $\tau_c$. Predictably, the thermal wave results will become independent to $\tau_c$ when $\tau_R \to \infty$.

The other case is the diffusive limit where $\tau_R$ is very small, for example 0.5 $\mu$s. Although the change of $\tau_c$ does introduce the appearance of the first sound peak, this peak carries a negligible
amount of energy, compared with the strong and heavy diffusive tail. Thus, it can hardly affect the heat transfer and become detectable. In addition, the small peak width of the first sound peak also causes technical difficulties for an accurate measurement. Similar to the other limit, the ballistic transport, the varying $\tau_c$ negligibly contributes to the heat transfer though a first sound peak better fits the physical point of view.

In sum, the EG-BTE is able to capture the features of second sound because of the second order derivative terms characterized by $\tau_c$. Parametric study shows that the ballistic effects are enhanced due to these terms, which essentially leads to the appearance of second sound, coinciding with its previous definition. Such a transitional phenomenon may significantly contribute to the nondiffusive thermal transport at room temperature, and the successful description of it by the EG-BTE may be the reason for the EG-BTE’s accurate predictions on multiple nondiffusive thermal transport experiments.

Next, the EG-BTE results are validated by the comparison to the published thermal wave experiments.

### 5.6.5 Validation with experiments

Previous experimental studies mainly focused on the temperature-dependence of the thermal wave signal. It is found that the transitional behavior, such as the second sound, is also sensitive to the temperature. We here propose the hypothesis that the temperature-dependence of thermal signal is due to the temperature-dependence of the parameters, $\tau_R$ and $\tau_c$, in the EG-BTE. As a consequence, both $\tau_R$ and $\tau_c$ have to be extracted from the experimental signal for each comparison case. Besides, the dependence of $\tau_R$ and $\tau_c$ on polarization modes is also considered, so that four parameters $\tau_{R,l}$, $\tau_{R,t}$, $\tau_{c,l}$ and $\tau_{c,t}$ are to be determined.

Fortunately, at first, the signal reveals clear separation of longitudinal and transverse compo-
Figure 5.21: The EG-BTE simulation of the transverse component of the thermal wave for small relaxation time $\tau_R = 0.5 \mu$s. The diffusive tail appears due to the intense scattering. There is still a sign of the second peak though obscured by the diffusive tail.
ments. The interaction between polarization modes are arguably not considered. A weighted superposition of longitudinal and transverse signal calculated independently is applicable. The weighting coefficient for the superposition is approximately 1 for the longitudinal component and 2 for the transverse component, assuming that the density states of these three modes are evenly distributed. Although there exists small disturbance around the exact number possibly due to the experimental noise, it does not harm the revealing of physics here. Moreover, due to the simplification, many rigorous physical details are not involved, such as phonon dispersion, different mechanisms of phonon interactions and so on. Thus, the comparisons below are just qualitative comparison. A quantitative study is left for future research.

![Graph](image)

Figure 5.22: Comparison between the EG-BTE results and experiments by Jackson et al. [54] at 9.6ºK. Clear two peaks of temperature pulses are detected for two polarizations of first sound.

### 5.6.5.1 Ballistic propagation

Prepared with the statements above, a ballistic signal experimentally reported by Jackson et al. [54] is regenerated by the EG-BTE using phonon $S_N$ method in Fig. 5.22. At this relatively low temperature, the relaxation time remains large and phonon scattering is low in intensity. Both longitudinal and transverse first peak are clearly observed and successfully reproduced by the EG-BTE. The fitted parameters are $\tau_{RI} = 1.4 \mu s$ and $\tau_{cl} = 0.42 \mu s$ for the longitudinal component, and $\tau_{RI} = 1.5 \mu s$ and $\tau_{cl} = 0.75 \mu s$ for the transverse component. The fitted values are very close to the $\tau_R = 1.56 \mu s$ given by Ma [58] calculated based on the assumption that, boundary scattering due to the Casimir limit [7] is dominant. From another perspective, the thermal conductivity at this temperature is roughly $\kappa = 8709.6$ W/mK given in Fig. 1 curve B [54]. With this measured thermal conductivity, $\tau_R$ can be qualitatively estimated through the definition of thermal conductivity:

$$\tau_R = \frac{3\kappa}{c_v c_s^2}$$  \hspace{1cm} (5.33)
where $c_v$ is the volumetric specific heat and $c_s$ is the Debye velocity related to the longitudinal velocity $c_l$ and transverse velocity $c_t$ by $c_s^{-3} = \frac{1}{3} c_l^{-3} + \frac{2}{3} c_t^{-3}$ [61]. According to Harrison et al. [62], the volumetric specific heat of NaF can be related to temperature as

$$c_v = 234Nk_B \left( \frac{T}{\Theta_D} \right)^3,$$

(5.34)

where $N$ is Avogadro’s number, $k_B$ is Boltzmann constant, $\Theta_D = 466 \pm 5^\circ$K is Debye temperature, $T$ is the temperature of interest.

Based on these formulas and estimated density of NaF $\rho \approx 2.6 \text{ g/cm}^3$, $\tau_R$ here is calculated as $\tau_R = 1.957 \mu$s. The fitted $\tau_R$ is also close to this number in magnitude. The fitted $\tau_c$, however, has no reference value to compared with. Fortunately, as previous stated, the thermal wave here is not very sensitive to the value of $\tau_c$ since the ballistic behavior is already strong.

5.6.5.2 Diffusive propagation

![Figure 5.23: Comparison between the EG-BTE results and experiments by Rogers et al. [55]. Strong diffusive behavior buries the first sound peaks.](image)

Rogers et al. [55], on the other hand, reported the thermal wave experiment results close to the other limit: diffusive transport. They provided not only the experimental data but also a computational model for simulating the thermal wave. The two results are together plotted in Fig. 5.23, along with the thermal wave calculated from the EG-BTE using phonon $S_N$ method. The EG-BTE, in this case, also predicts a correct trend of the thermal wave, which suggests its proved ability to physically model thermal transport even in the low temperature case. The extracted parameters are $\tau_{Rl} = 0.007 \mu$s for the longitudinal component, and $\tau_{Rt} = 0.03 \mu$s for the transverse component. No $\tau_c$ for either component can be fitted since the result is already insensitive to $\tau_c$ again, as mentioned previously.
A reference value of $\tau_R$ can be obtained by conducting similar calculations starting from the thermal conductivity of NaF given in Fig. 11 [55]. According to this figure, thermal conductivity at this temperature ranges from 2000 W/mK to 6000 W/mK, indicating an overall $\tau_R$ from 0.024 $\mu$s to 0.072 $\mu$s. Although our extracted $\tau_{Rl}$ falls out of bounds, $\tau_{Rt}$ fits right in, possibly implying more weight of energy carried by the transverse component.

However, here, one may notice that Roger’s numerical results almost exceeds the upper noise bound at the tail, while our simulation fits right into the central zone between the two bounds. Actually, Ma [58] reported similar trend of the tail behavior. This minor discrepancy is most possibly due to the energy loss through the interface between the sample and the thermometer. The thermometer receives and absorbs a small amount of energy, equivalently cooling down the backside of the sample cube. In the ballistic case, no such cooling effect is accounted because of the short duration time monitored. However, as the duration of the experiment observation increases, this cooling effect introduces differences. To qualitatively capture this and better model the practical situation, we implement the Robin type boundary condition given by Eq. (3.38) in our simulation, to physically reproduce the heat transfer through the interface to the thermometer.

Having been validated by the ballistic and diffusive limits, the EG-BTE is compared to the transitional behavior in the following part.

5.6.5.3 Transitional propagation

McNelly et al. [5] reported the heat pulses experiments in NaF with the second sound observed. Fig. 1 in their paper clearly illustrates this transitional phenomenon, and thus selected to compare with the EG-BTE. The results are shown in Fig. 5.24 without a numeric description on the amplitude. McNelly et al. explained that, the pulse amplitudes cannot be compared for different traces, because the amplifier gain was increased with increasing temperature. The extracted parameters are specified in Table 5.3. McNelly et al. mentioned that the maximum thermal conductivity $\kappa_{\text{max}} = 7000$ W/mK was measured at 12°K for the experiment sample [5]. As a result, a maximal $\tau_R = 0.805$ $\mu$s can be calculated using Eq. (5.33) at 12°K. Overall, our fitting does qualitatively reveal the trend of $\tau_R$ changing with temperature. The discrepancy that our maximal $\tau_R$ shows up at 11.0°K instead of 12°K may be due to the effects of $\tau_c$.

At different environmental temperatures, thermal wave propagation performs differently. The ballistic transport similar to Fig. 5.22 is observed at 7.8°K on the top, and the diffusive transport similar to Fig. 5.23 is observed at 14.2°K at the bottom, though it is not as purely diffusive as in Fig. 5.23. The transitional transport is shown by the two curves in the middle, where the second sound broadens the first sound of transverse wave significantly in an abnormal fashion. The second sound itself does not appear completely as a peak, but a small emerging dune can be found in the thermal wave trace at 12°K (blue line). It can be inferred based on the parametric study that this small dune may grow into the second sound peak once the redistribution of phonon energy is strengthened by some treatments on the sample that improves $\tau_c$.

In the bottom case, unfortunately, not all features are successfully regenerated by the EG-BTE, such as the second sound peak by the longitudinal wave (though the EG-BTE does have one). The EG-BTE is still able to predict a correct tendency even if it does not closely follow the experimental signal. Further study is needed here to explore the thermal transport in this regime.

There is another experiment case by McNelly et al. with stronger effects from second sound. However, the EG-BTE cannot reproduce the transitional behavior in this case. This is possibly
Figure 5.24: Comparison between the EG-BTE results and experiments by McNelly et al. [5]. A transition from ballistic to quasi-diffusive transport is captured at different environmental temperatures.
because the purity of the crystal applied in the experiment is so high that the different scattering mechanisms are necessary to be taken into account. If so, the single relaxation time assumption fails. Moreover, the implementation of phonon dispersion should facilitate to better describe the transport mechanisms here.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Temperature</th>
<th>$\tau_{Rl}$</th>
<th>$\tau_{cl}$</th>
<th>$\tau_{Rl}$</th>
<th>$\tau_{cl}$</th>
<th>$\tau_R$ from $\kappa$</th>
<th>Regime</th>
</tr>
</thead>
<tbody>
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<td>[54]</td>
<td>9.6°K</td>
<td>1.4</td>
<td>0.42</td>
<td>1.5</td>
<td>0.75</td>
<td>1.96</td>
<td>Ballistic</td>
</tr>
<tr>
<td>[5]</td>
<td>7.8°K</td>
<td>1.4</td>
<td>0.31</td>
<td>1.3</td>
<td>1.3</td>
<td>N/A</td>
<td>Ballistic</td>
</tr>
<tr>
<td>[5]</td>
<td>11.0°K</td>
<td>1.5</td>
<td>0.20</td>
<td>1.7</td>
<td>0.75</td>
<td>N/A</td>
<td>Transitional</td>
</tr>
<tr>
<td>[5]</td>
<td>12.0°K</td>
<td>1.5</td>
<td>0.20</td>
<td>1.3</td>
<td>0.75</td>
<td>N/A</td>
<td>Transitional</td>
</tr>
<tr>
<td>[5]</td>
<td>14.2°K</td>
<td>0.5</td>
<td>0.18</td>
<td>0.32</td>
<td>0.09</td>
<td>N/A</td>
<td>Diffusive</td>
</tr>
<tr>
<td>[55]</td>
<td>25.5°K</td>
<td>0.007</td>
<td>N/A</td>
<td>0.03</td>
<td>N/A</td>
<td>0.048</td>
<td>Diffusive</td>
</tr>
</tbody>
</table>

Table 5.3: Summary of fitted $\tau_R$ and $\tau_c$. The $l$ and $t$ in parameter subscript represents longitudinal and transverse, respectively. The unit of all numeric values is $\mu$s. Calculated results using Eq. 5.33 are also listed as references in the second last column.

5.6.6 Size- & temperature-dependence of thermal wave

In last section, the increase of temperature is experimentally found to initiate the transition from ballistic transport to the diffusive transport. In addition to this well-recognized temperature dependence, McNelly et al. also pointed out that the second sound is insensitive to the heating pulse width $t_p$, but sensitive to the path lengths. Although the size-dependence has been found in early days, no much attention was paid to its impact on thermal transport. This statement, however, does imply that an appropriate size range is needed for the observation of second sound.

Having successfully reproducing the experimental results, the EG-BTE is implemented to study the size-dependence of the thermal wave phenomenon. By simply changing the total length of the computational domain, the thermal wave signals on different sample sizes can be generated. Another option to study the propagation is to sample the temperature history at several selected cross-sections, but such a method excludes the effects from the boundary and thus is not taken for the first case. Here, we select four sizes of the cube to analyze the size-dependence of thermal wave propagation and its similarity to temperature dependence, and the parameters $\tau_R$ and $\tau_c$ are the fitted values based on experimental results. During the simulation, only the transverse component is simulated. This is because the final signal is simply a superposition of the two components, and the second sound mainly falls in the transverse component. Moreover, due to the shrinking in size, these two first sound peaks are overlapped, obscuring the analysis if superposition is performed. With these specifications, the simulation results are shown in Fig. 5.25.

According to the simulation results, the ballistic transport can be observed as a clear first sound peak with the NaF sample size $L = 2.34$ mm, which is similar to the transverse first sound peak in the trace at 7.8°K. As the size increases to $L = 3.9$ mm, a transitional transport without second sound similar to the experimental trace at 11.0°K can be found. Next, a clear second sound peak can be observed with modified size $L = 7.02$ mm even though it is buried in the experimental signal. At last, when the size reaches $L = 11.7$ mm, the calculated trace shows similarity with the quasi-diffusive case at 14.2°K, as the thermal wave propagation reaches well-developed stage.
In addition to this experimental case, another case by Rogers et al. [55] is also studied by monitoring the temperature history at several locations to reveal the development of thermal wave during propagation, as given in Fig. 5.26. Both longitudinal and transverse components are calculated and exhibited here. In this diffusive dominant case, however, ballistic transport can be observed at a cross-section $x = 0.1L$. Longitudinal component is already dissipated since $\tau_{RL}$ is very small. The transverse one, on the other hand, still performs ballistically with a second sound peak. This second sound peak is weak because the majority of energy is still traveling as the first sound. Later, at the next cross-section $x = 0.2L$, second sound peak is broadened to a strong tail. This tail closely follows the first sound peak as a collection of the scattered phonons from the first peak. As the offset distance from the heating surface grows to $x = 0.4L$, the first sound peaks turns almost undetectable, marking that the ballistic behavior has already been dissipated. Finally, a strong and heavy diffusive tail is full-grown with no oscillation standing for the ballistic transport, or wave-like behavior.

Such a propagation developing process is possible to be undergoing in the vicinity of heating source at room temperature. This could explain from the view of physics why EG-BTE is capable of predicting the size-dependence ETC in mesoscale. Based on this, the physical meaning of $\tau_c$, the characteristic time describing nonlocal effects of ballistic phonon transport, can be better explained as a signal of phonon energy redistribution phenomenon. As suggested by its name, this characteristic time contributes to ballistic phonon transport.

At last, it should be noted that the similarity between size-dependence and temperature-dependence, intuitively, suggests a relation between $\tau_R$, $\tau_c$ and the size $L$ for thermal wave propaga-
Figure 5.26: Transitional behavior observed at different cross-sections of the sample during simulation for experiments by Rogers et al. [55]. Size-dependent transition from ballistic regime to diffusive regime is predicted by the EG-BTE.
tion, or even more general, for heat transfer. The effect by $\tau_c$ is not as radical as temperature, which directly causes the change of both $\tau_R$ and $\tau_C$, but it could possibly be linked to the size-dependence, and provide more hint for a more systematic study on this direction.

5.6.7 Observing thermal wave at room temperature

![Figure 5.27: The thermal wave on silicon sample predicted by the EG-BTE at room temperature. All material properties including $\tau_R$ and $\tau_c$ are from 1D and 2D TTG. The maximal temperature rising over the whole process is set to be 1.](image)

In the previous sections of this chapter, the material parameters have been fitted for bulk silicon material. Since our goal is to predict the observation of the thermal wave at room temperature, we can perform the simulation on a bulk silicon cube with the same setup as at low temperature. Thus, an example simulation is carried out. In this example, the thickness of the material is 10 $\mu$m to make sure there is no reflection from the boundary from the backside, which is considered insulated. On the heating side, a Gaussian shaped pulse heating with half peak width $\Delta t = 20$ ps is implemented at the moment when $t = 0$. Different from what was implemented in the thermal wave experiments at low temperature, the heating here is a heat flux deposited on the surface of the silicon sample.

Figure 5.27 shows the temperature history at different locations of the sample. Specifically, $x = 0.0 \mu$m illustrates the surface temperature history. It show a clear oscillatory behavior, which is essentially different from the diffusive or nondiffusive phenomena, where the temperature monotonically drops in time. This result also indicates the amplitude of the oscillatory behavior decays into the depth, just as we observed in the thermal wave experiment at low temperature: this peak gradually dissipates during propagation.

According to Fig. 5.27, the heat pulse in bulk silicon at room temperature remains detectable (amplitude decays no more than 90%) within the traveling distance of 0.6 $\mu$m. Although this length scale is technically not hard to observe nowadays, unfortunately, this ultrafast heating is hard to obtain. Joule heating may not be able to reach such a frequency and sensitivity, while laser heating
may introduce the SAWs that contaminate the signal, making the source of oscillation indistinguishable. Furthermore, the surface temperature can be measured similar to the TTG techniques, but the temporal resolution has to be pretty high (i.e., 50 ps) to distinguish the temperature peak. But overall, the EG-BTE suggests such a possibility for a direct observation of thermal wave at room temperature when the experiment is delicately designed.

5.6.8 Discussion

At the end of this section, we have some thoughts to address regarding the details of this study.

**Phonon dispersion & scattering mechanism** The definition of second sound according to McNelly et al. is related to the momentum-conserving scattering of phonons. Thus, distinct scattering mechanisms should be separately treated instead of simply merging into one average relaxation time $\tau_R$. This could explain why some experimentally observed features cannot be reproduced by the EG-BTE. Moreover, the phonon dispersion is another indispensable input if more physics is to be included. The phonon dispersion introduces multiple group velocities of phonons of different frequencies, and finally will lead to the modification on the waveforms and the merging of second sound peak. In addition to the two second derivative terms in the EG-BTE, the phonon dispersion is possible to be an important or dominant factor in the simulation of transitional transport.

![First order and second order upwind scheme](image)

Figure 5.28: First order and second order upwind scheme. The red solid line shows the results by first order scheme, while the green dashed line represents the results by second order scheme. Strong dissipation is observed for the first order result (the removal of the first peak), and nonphysical disturbance is introduced by the second order result (numerical oscillation on the tail).

**Numerical error** The phonon $S_N$ method requires certain numerical algorithm to solve the discretized direction-dependent equations. Here, due to the fast heating, the problem’s stiffness
in time brings about numerical issues, as illustrated by Fig. 5.28. Since finite difference method is applied to discretize EG-BTE in time and space, the method unavoidably shares disadvantages as discovered in traditional Computational Fluid Dynamics study. Fig. 5.28 describes an extreme case. First order upwind scheme exhibits strong numerical dissipation to the degree where the first sound peak disappears, while second order upwind scheme brings about strong dispersion. In addition to this numerical issue by finite difference method, the discretization of solid angle also adds to the numerical oscillation.

Traditionally, there are several ways to damp the numerical oscillation, such as the WENO method [63], the application of artificial viscosity term [64], the finite analytical method [65]. But all the results shown in this chapter have already been tested by matching the results from first and second order method. Furthermore, grid-independence has been checked as well.

5.6.9 Conclusion for thermal wave experiments

In this section, thermal wave propagation at low temperature has been successfully simulated using the EG-BTE solved by phonon $S_N$ method. All three transport regimes, ballistic, transitional and diffusive regimes, have been reproduced and validated by comparison with experimental results. The second sound captured in transitional transitional regime is also regenerated by the EG-BTE, proving its ability to offer correct prediction in this regime. Through this agreement with experiments at low temperature, the EG-BTE unifies the distinct thermal transport behaviors at low temperature and room temperature, bridging the heat transfer at different temperature ranges. Besides, it also suggests the observation of thermal wave in a desirable length scale (approximately 1 µm), though some technical difficulties exist.

5.7 Summary

In this chapter, the Enhanced Gray Boltzmann transport equation (EG-BTE) is applied on 1D/2D transient thermal grating (TTG) experiments, metallic grating experiments (MG-X-ray experiments) and thermal wave experiments at low temperature.

In 1D/2D TTG experiments, the analytical solutions of the EG-BTE give very similar results as by the TPHC model, validating the derivability from the EG-BTE to the TPHC model. Non-diffusive heat transfer found in the experiments is also explained by the EG-BTE. Furthermore, the EG-BTE is extensively implemented on artificial cases and compared with the gray BTE and spectral BTE.

In MG-X-ray experiments, the numerical solution of the EG-BTE by lattice Boltzmann method (LBM) is validated by the MG-X-ray experimental results. Due to EG-BTE’s validated ability to appropriately describe the nondiffusive heat transfer inside the substrate, the size-dependence of the interface resistivity has been successfully extracted. Based on this size-dependence, a hypothesis is proposed regarding the nondiffusive heat transfer across the interface.

In thermal wave experiments at low temperature, the in-house numerical scheme, the discrete ordinates method for phonon transport (phonon $S_N$ method) is validated and applied to solve the EG-BTE in this situation. Validated by comparison with the experiments, the EG-BTE shows the potential to cover the full range from the ballistic regime to the diffusive regime, bridging the gap between the atomic level tools and the diffusive model.
In addition, the possible existence of ballistic phonon transport is discussed using the EG-BTE in all these experiments to reveal more hidden physics and benefit the fundamental study.

To sum up, the EG-BTE is validated by multiple nondiffusive heat transfer experiments. It reveals hidden physics about the ballistic phonon transport, which is a new regime that has not been experimentally discovered. Its wide range of applicability not only supports the validity of the TPHC model, but also improves our understanding of heat transfer from the perspective of fundamental study.
Chapter 6

Summary and Future Work

6.1 Concluding Remarks

This study aims at both engineering-assisting tool and exploration of fundamentals for nondiffusive heat transfer. Here, we summarize this study from the two perspectives.

6.1.1 Practical remarks

The engineering objective of this thesis is to provide a tool for describing nondiffusive heat transfer in dielectric materials equipped with both solid reliability and low computational expense. This tool should satisfy three major requirements: (1) it is a validated nondiffusive heat transfer models that explain the physics of phonon transport in nano-/meso-scale and provide convincing prediction on the thermal transport; (2) it should come with efficient numerical tools that implement the model with acceptable simplicity and accuracy for engineering design applications; (3) it offers a criterion for a qualitative evaluation on the validity of Fourier’s law to guide whether nondiffusive models are necessary in a given engineering situation to lower the computational cost. In this thesis, this tool has been found, that is, the Two-Parameter Heat Conduction model.

In this thesis, first, the analytical solutions of the TPHC model are developed and validated on 1D and 2D transient thermal grating experiments, improving the efficiency for experimental data analysis. Second, in addition to the analytical solutions, the numerical solutions of the TPHC model through a regular finite difference method can also capture the nondiffusive phenomena in the MG-X-ray experiments and allow the extraction of a clean effective interface resistivity without the impacts from the nondiffusive heat transfer inside the substrate. Third, the nondimensional \( \zeta \) defined based on the TPHC model offers a quick evaluation on the applicability of Fourier’s law in given engineering situation, serving as a criterion to determine whether nondiffusive models are required in this situation.

6.1.2 Theoretical remarks

Besides potential practical application, the physical objective of this study is to offer more insights on the fundamentals of nondiffusive heat transfer either inside the materials or across the interface.
Supported by multiple experimental observations, EG-BTE is capable of covering all three different heat transfer regimes up to diffusive and down to ballistic. This unified model also offers explanation on the seemingly anisotropic heat transfer in multidimensional situation. In addition, it also predicts the appropriate time and length scale for the direct measurements of ballistic phonon transport, opening up the possibility for deeper and more comprehensive understanding of heat transfer fundamentals.

Besides, the size-dependence of effective thermal interface resistivity due to the nondiffusive heat transfer across the interface has also been studied by the EG-BTE. Based on the extracted interface resistivity from the experiment, EG-BTE inspires the hypothesis that the effective interface resistivity mainly depends on its own size. This hypothesis, though remains to be further validated, contributes to the fundamental research of the thermal transport across the interfaces and suggests further guidance for experimental studies.

6.2 Future Work

Although lots of researches have been done in thesis, more possible directions have been open for future work. We here address some aspects of future study.

6.2.1 TPHC model and corresponding boundary conditions

The TPHC model requires one more boundary conditions, as stated in Chapter 4. However, if the last two terms in Eq. (2.11) can be combined as one term, just as in 1D situations, no extra boundary condition is needed. This mathematically means irrotational behavior for heat flux \( \frac{\partial q}{\partial y} = \frac{\partial q}{\partial x} \), as commonly believed. However, the existence of “heat vortex”, though seems irrational from conventional thoughts, still needs rigorous falsification from the physical point of view. Thus, TPHC model is complete with both governing equations and boundary conditions determined.

On the other hand, due to its mathematical simplicity and compatibility with the EG-BTE, the TPHC model shows great potential on improving the accuracy of simulations for engineering use while maintaining an affordable computational costs. Its applications should also be extended to practical engineering cases for more validation, and hopefully serves as a replacement of Fourier’s law in the situations where nondiffusive effects, characterized by the nondimensional parameter \( \zeta \), are significant.

6.2.2 Refinement of numerical tools

In this thesis, the present equilibrium distribution function in highly nonequilibrium situations shows significant departure from the analytical solutions. Although the other numerical tool provided in this thesis, phonon \( S_N \) method, does not suffer from this issue, LBM still naturally has advantages in efficiency over phonon \( S_N \) method. If this problem can be eliminated, the computational efficiency of the EG-BTE can be considerably improved.

Besides LBM, phonon \( S_N \) method so far has only been implemented and validated in 1D situations. Its extension on multidimensional cases can possibly lead to more physical results and details.
6.2.3 Phonon transport described by the EG-BTE

In Chapter 5, a qualitative study on thermal wave experiments has been done, and prediction for the direct measurement of phonon propagation as thermal waves is proposed. However, a quantitative study including the information of the thermal wave peak amplitudes will undoubtedly offer more insights on the dissipation of the ballistic phonon transport and the energy redistribution leading to the second sound. Since second sound is an important feature of transitional phonon transport, this quantitative study can also provide implication on the size-dependence of heat transfer at room temperature.

In thermal wave study, the similarity between size-dependence and temperature dependence also implies the existence of a nondimensional parameter to link them together, just as $\zeta$ for the TPHC model. The discovery of this parameter can predictably leads to the characterization of phonon transport regimes, and serves as a reference for model selection in calculation.

6.2.4 Nondiffusive heat transfer model for thermal interface

At present, the concept of effective interface resistivity is still constructed under the framework of Fourier’s law. Although nondiffusive heat transfer models have been attracting researchers for several decades, no reliable nondiffusive interface model has been publicly recognized. This model, once developed and validated, may point us to the origin of size-dependence of interface thermal transport, and thus completes the theory of nondiffusive heat transfer.
Bibliography


