A Generalized Enhanced Fourier Law

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A generalized enhanced Fourier law (EFL) that accounts for quasi-ballistic phonon transport effects in a formulation entirely in terms of physical observables is derived from the Boltzmann transport equation. It generalizes the previously reported EFL from a gray phonon population to an arbitrary quasi-ballistic phonon mode population, the chief advantage being its formulation in terms of observables like the heat flux and temperature, in a manner akin to the Fourier law albeit rigorous enough to describe quasi-ballistic phonon transport. [DOI: 10.1115/1.4034796]

1 Introduction

Reports of significant room-temperature quasi-ballistic phonon transport [1–3] have spurred modeling efforts [4–11] aimed at explaining observed experimental results and predicting new effects. Each approach has specific advantages in certain situations, resulting in a wide variety of mathematical formulations. The aim of this paper is to derive a useful Fourier-like formulation, which we term the generalized enhanced Fourier law.

Few analytical or semi-analytical phonon conduction models exist that treat the full nonlinear Boltzmann transport equation (BTE) [12]. Fully numerical solutions are outside the purview of this paper. All models discussed here assume a reference temperature and hence a reference equilibrium Bose distribution function, and assume the validity of the linearized BTE under the relaxation-time approximation.

This paper is organized as follows: Section 2 surveys the current literature on quasi-ballistic transport models; Sec. 3 derives the generalized EFL and notes its correspondence with the seminal work of Maznev et al. Section 4 summarizes our discussion.

2 Brief Survey of Major Quasi-Ballistic Transport Frameworks

2.1 The Enhanced Fourier Law. Ramu et al. [8] proposed a technique to arrive at the heat flux of a quasi-ballistic mode directly from the BTE by truncating the spherical harmonic expansion of the distribution function at the $l = 2$ order in angular momentum. Since the heat flux is a physically more accessible quantity than the distribution function, this formalism has certain advantages over others, some of which are (a) the angular integrals over distribution functions have already been performed, and nonlocality of the quasi-ballistic heat flux emerges naturally, (b) energy conservation is easier to enforce, and (c) modal suppression functions may easily be derived, as exemplified in Sec. 3.

2.2 Chen's Ballistic-Diffusive Equations and Series Solution of Ordonez-Miranda et al. Chen's ballistic-diffusive equations [6] (BDEs) and Ordonez-Miranda et al. [7] recognize that the homogenous Boltzmann equation is a damped advection equation, to which a closed-form solution exists. For the inhomogeneous solution, while the BDE assumes the Fourier law, the solution of Ordonez-Miranda et al. assumes a gray medium (constant relaxation time for all modes) and expands the BTE solution in a Taylor series in the spatial coordinate to arrive at a beyond-Fourier constitutive law for the quasi-ballistic heat flux.

2.3 Weakly Quasi-Ballistic Solution of Maznev et al. and Generalized BTE Solution of Hua and Minnich. In the context of the transient grating experiment, Maznev et al. [4] presented an exact BTE solution by taking the Fourier transform and writing the set of coupled BTEs for each mode as an eigenvalue equation, which was solved for the transient grating decay time. By comparing the decay time with the Fourier law prediction, a correction factor called the "suppression function" was derived, from which the mean free path accumulation function [13] could be recovered via a reconstruction technique [14].

Hua and Minnich [5] subsequently solved the full BTE semi-analytically with an improved energy conservation equation that takes into account the nonequilibrium modes instead of the usual procedure of ignoring them and lumping the equilibrium modes into a heat-capacity term.

They found two distinct transport regimes, namely, the weak and strong quasi-ballistic regimes, depending on the relative magnitudes of phonon-mode scattering lifetime and the overall thermal decay time. Upon assuming the modal decay time to be much less than the thermal time constant for decay of the transient grating, the solution of Maznev et al. automatically corresponds to the weakly quasi-ballistic limit of Hua and Minnich.

2.4 The Models of Regner et al., Yang and Dames, and Maassen and Lundstrom. Regner et al. [15] have proposed a gray model (constant mean free path and constant velocity for all modes). They truncate the spherical harmonic approximation of the distribution function at the lowest nontrivial order, namely, $l = 1$ as opposed to $l = 2$ of the enhanced Fourier law. In fact, except for different boundary conditions, their formulation is essentially the Cattaneo equation, as can be seen by introducing the equilibrium Bose distribution in Eq. (7*a*) of that work and summing over all modes. However, two counterpropagating heat fluxes are separately solved for, which compensates for the low order to which each is analyzed. The gray assumption makes it difficult to compare with other approaches.

Yang and Dames [16] have used the Milne–Eddington approximation similarly to Ref. [15]; however, they have generalized their treatment to a nongray population. Here again, introduction of forward and reverse propagating heat fluxes compensates for the low order of spherical harmonic expansion. But for the way that the anisotropy of phonon population in *k*-space is handled, the approach of Yang and Dames is similar to the EFL. Their formulation has one advantage, namely, that boundary conditions may be easily prescribed for one planar surface.

Maassen and Lundstrom [17–19] have utilized the McKelvey–Shockley approach to derive a formulation of the BTE where the central quantity is the heat flux. This heat flux is divided into two counterpropagating components and the boundary conditions are prescribed for each component. This formulation is capable of describing highly nonequilibrium transport; however, it has only been studied for 1D transport between perfectly absorbing (reflection-less) thermal contacts.

3 The Generalized Enhanced Fourier Law

The enhanced Fourier law (EFL), as presented previously in Ref. [8], handles quasi-ballistic modes of a single mean free path. It has utility in explaining the transient grating [20] and the

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frequency-domain thermoreflectance experiments [10,11]. Here, we generalize it to handle arbitrary mean free path (MFP) spectra. To do so, we track the original derivation of Ramu [8] until the assumption of constant mean free path is made. The generalization is necessary for meaningful comparison with the equations of Maznev et al. who used a Fourier transform in both time and space to reduce the set of BTEs for the phonon modes to an eigenvalue equation. The following discussion additionally derives a generalized EFL, a differential equation for heat transfer by modes of multiple MFPs. This is of vital importance to realistic thermal modeling of silicon devices, because crystalline silicon has a wide spectrum, with phonons of about three orders of magnitudes in MFPs contributing significantly to thermal transport. Although in this section we will be focusing on 1D transport, Appendix A gives the equation in three dimensions, where a new circulatory term (term with nonzero curl) arises. Furthermore, although Fourier transforms in both spatial and temporal variables will be used throughout the development so as to deal with purely algebraic equations, the resulting expressions will be rational polynomials in the transformed variables, and as such the inverse transform may easily be taken to yield corresponding partial differential equations.

The basis of the EFL is the assumption that quasi-ballistic modes do not interact with each other due to the small phase-space for such scattering [4], but can exchange energy with the reservoir, which is assumed to exist at temperature T. We denote the distribution function for quasi-ballistic modes as $g(x,\boldsymbol{k})$, where all spatial variation is assumed to be along the x-direction, and $\boldsymbol{k}$ is the phonon-mode wavevector of magnitude k and making an angle θ with the x-axis.

We expand the distribution function in terms of spherical harmonics $P_l(\cos\theta)$. Spherical harmonics form a complete, orthogonal basis for expanding angle-dependent azimuthally symmetric functions [21]

$$g(x,\boldsymbol{k},t) = \sum_{l=0}^{\infty} g_l(x,k,t) P_l(\cos\theta) \tag{1}$$

For the sake of brevity, we henceforth suppress the $(x,k,\ t)$ dependence wherever no ambiguity arises. Also, here and henceforth, symbols in bold fonts denote vectors, while the same symbols in normal fonts denote their magnitudes.

We first derive the equation for phonon modes of wavevector magnitude k, lifetime $\tau(k)$, and group-velocity magnitude $v(k)$. The frequency of a quasi-ballistic mode of wavevector $\boldsymbol{k}$ is denoted by $\omega(k)$. We also assume isotropic phonon dispersion, so that $\boldsymbol{v} = v(\boldsymbol{k}/k)$. The time-dependent linearized BTE for the quasi-ballistic modes is then given by

$$v\cos\theta \frac{\partial g(x,\boldsymbol{k},t)}{\partial x} + \frac{\partial g(x,\boldsymbol{k},t)}{\partial t} = -\frac{g(x,\boldsymbol{k},t) - f_{\text{Eq}}(x,k,T)}{\tau} \tag{2}$$

We have earlier assumed the existence of a local temperature T and thereby a corresponding spherically symmetric equilibrium distribution, familiar from Bose statistics: $f_{\text{Eq}}(x,\boldsymbol{k},T) = f_{\text{Eq}}(x,k,T) \equiv f_{\text{Eq}}(T)$. This local temperature is established by some high-capacity reservoir. Equation (2) is the quasi-ballistic mode BTE in the absence of source terms; thus, we assume that no external source of heat couples to the quasi-ballistic modes. We begin with the observation that, owing to the orthogonality of the spherical harmonics [21], the x-component of the quasi-ballistic heat flux is determined solely by the first spherical harmonic g_1

$$\begin{aligned} Q(x,t) &= 2\pi \sum_k \int_{\theta=0}^{\pi} \hbar\omega g(x,\boldsymbol{k},t) v\cos\theta \sin\theta d\theta \\ &= \frac{4\pi}{3}\sum_k \hbar\omega v g_1(x,k,t) \end{aligned} \tag{3}$$

This may be seen by substituting Eq. (1) for $g(x,\boldsymbol{k},t)$ in the integrand and applying orthogonality of the spherical harmonics. Specifically, $\cos\theta$ is the $l=1$ spherical harmonic, and since all other spherical harmonics are orthogonal to it, only the $l=1$ term survives. Therefore, we seek a differential equation for g_1. Here and henceforth, $\sum_k I(k)$ is shorthand for $[1/(2\pi)^3]\int dk I(k) k^2$ where $I()$ is any function of k, and the integral is over all quasi-ballistic mode wavevector magnitudes.

Highly nonequilibrium transport in electron gases has been investigated analytically by Baraff [22]; we adopt his approach. First, we take the Fourier transform in time of Eq. (2), with transform variable γ. Substituting Eq. (1) into the Boltzmann transport equation (Eq. (2)), multiplying successively by $P_{l'}(\cos\theta)\sin\theta$ for $l' = 0,1,2,\ldots$, and integrating over θ, we arrive at a hierarchy of coupled equations for the g_l s [22]; the first three of which are

$$\frac{1}{3}v\frac{\partial g_1}{\partial x} + \frac{g_0}{\tilde{\tau}} - \frac{f_{\text{Eq}}(T)}{\tau} = 0 \tag{4a}$$

$$\frac{2}{5}v\frac{\partial g_2}{\partial x} + v\frac{\partial g_0}{\partial x} + \frac{g_1}{\tilde{\tau}} = 0 \tag{4b}$$

$$\frac{3}{7}v\frac{\partial g_3}{\partial x} + \frac{2}{3}v\frac{\partial g_1}{\partial x} + \frac{g_2}{\tilde{\tau}} = 0 \tag{4c}$$

Here, $\tilde{\tau}$ is given by $(1/\tilde{\tau}) = (1/\tau) + j\gamma$ where, as before, γ is the Fourier transform variable in time, and j is the imaginary unit. We truncate the hierarchy at the second order by setting $g_3 = 0$; other truncations are possible [22]. The generalization of Eq. (4) to arbitrary order in spherical harmonics is given in Appendix A. Therefore, this is the leading approximation beyond the Fourier law, which consists of setting $g_2 = 0$. Substituting Eq. (4c) into Eq. (4b) to eliminate g_2, and the result into Eq. (4a) to eliminate g_0, we arrive at an equation solely in terms of g_1

$$-\frac{3}{5}(v\tau)^2\frac{\partial^2 g_1}{\partial x^2} + v\tau\frac{\partial f_{\text{Eq}}(T)}{\partial x} + (1+j\gamma\tau)^2 g_1 = 0 \tag{5}$$

where $f_{\text{Eq}}(T)$ depends on x only through T, enabling the replacement

$$\frac{\partial f_{\text{Eq}}(T)}{\partial x} = \frac{\partial f_{\text{Eq}}(T)}{\partial T}\frac{\partial T}{\partial x}$$

We make the substitution here of $v(k)\tau(k) = \Lambda(k)$, the k-dependent MFP of quasi-ballistic phonons, to yield

$$-\frac{3}{5}\Lambda^2\frac{\partial^2 g_1}{\partial x^2} + \Lambda\frac{\partial f_{\text{Eq}}(T)}{\partial T}\frac{\partial T}{\partial x} + (1+j\gamma\tau)^2 g_1 = 0 \tag{6}$$

This is the point of departure from the original EFL. Taking the spatial Fourier transform $G_1(\chi,k,\gamma)$ of $g_1(x,k,\gamma)$ with transform variable χ replacing x, and rearranging, we get

$$G_1(\chi,k,\gamma) = \frac{-j\Lambda\dfrac{\partial f_{\text{Eq}}(T)}{\partial T}\chi T}{(1+j\gamma\tau)^2 + \dfrac{3}{5}\Lambda^2\chi^2} \tag{7}$$

Using Eq. (3), we arrive at

$$\begin{aligned} Q(\chi,\gamma) &= -j\chi T\int_{k=0}^{k_{\max}} \frac{\dfrac{1}{3}C(k)v(k)\Lambda(k)}{(1+j\gamma\tau)^2 + \dfrac{3}{5}\Lambda^2\chi^2}dk \\ &= -j\chi T\int_{k=0}^{k_{\max}} \frac{\kappa_{\text{diff}}(k)}{(1+j\gamma\tau)^2 + \dfrac{3}{5}\Lambda^2\chi^2}dk \end{aligned} \tag{8}$$

where the differential heat capacity with respect to k is $C(k) = [1/(2\pi)^3]4\pi\hbar\omega(k)[(\partial f_{\text{Eq}}(T))/\partial T]k^2$, and the differential thermal conductivity is $\kappa_{\text{diff}}(k) = (1/3)C(k)v(k)\Lambda(k)$. Equation

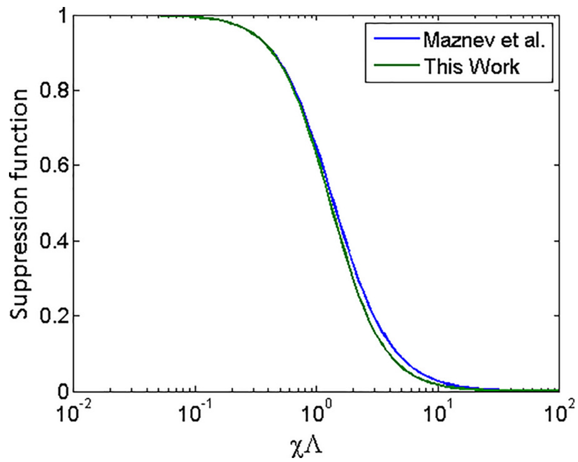

Fig. 1 Suppression functions of Maznev et al. [4] and this work (see Eq. (8)) match very closely over a large range of values of $\chi\Lambda$

(8) is equation of the generalized EFL for the Fourier transform of the net heat flux.

If we interpret χ as the inverse spatial period in the transient grating experiment (although this is unnecessary—see Appendix B), and restrict ourselves to such small phonon lifetimes that $\gamma\tau \ll 1$, it is seen that Eq. (8) is of the same form as Eq. (18) of Maznev et al. Specifically, the "correction factor" or suppression function of their work [4], $A_{\rm Maznev}(\chi\Lambda) = [3/(\chi^2\Lambda^2)]\{1-[(\arctan(\chi\Lambda))/\chi\Lambda]\}$ is replaced by $A_{\rm EFL}(\chi\Lambda) = 1/\{1+[(3/5)\ \Lambda^2\chi^2]\}$. Figure 1 compares the two functions for a broad range of values of $\chi\Lambda$, and it is seen that they match very closely. Since the work of Maznev et al. treats the exact two-channel BTE, the truncation to the second order of our spherical harmonic expansion gives very small errors indeed.

The main limitation of the EFL stems from the fact that it corresponds to a second-order spherical harmonic expansion of the phonon distribution function. Clearly by ignoring higher orders, it cannot describe the strong quasi-ballistic ballistic transport regime of Hua and Minnich. Thus, the EFL is not recommended for situations where the overall time constant of the experiment is expected to be on the order of the phonon lifetime. Finally, it is to be noted that the EFL is simply a constitutive equation for the heat flux. It is necessary to combine it with energy conservation and solve with appropriate boundary conditions to yield the temperature profile.

4 Conclusions

The enhanced Fourier law has been derived and generalized from a gray phonon population to an arbitrary one. The resulting suppression function for the effective thermal conductivity in the transient grating experiment has been shown to closely approximate results of Maznev et al. in the weakly quasi-ballistic transport regime. The chief advantage of the EFL is seen to be its formulation in terms of observables like the heat flux and temperature, akin to the Fourier law but rigorous enough to be capable of describing quasi-ballistic phonon transport. This feature is highly attractive in the context of simple explanations of quasi-ballistic transport experiments like the transient grating and frequency-domain thermoreflectance experiments, and is likely to promote physically accurate device thermal simulations.

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Nomenclature

A = suppression function
BDE = ballistic-diffusive equations
BTE = Boltzmann transport equation
$C(k)$ = differential heat capacity with respect to k
C_v = heat capacity of all phonon modes, J/kg K
EFL = enhanced Fourier law
$f_{\rm Eq}$ = Bose equilibrium distribution function
$g(x,\boldsymbol{k})$ = distribution function for quasi-ballistic modes
G = spatial Fourier transform of g
$\hbar$ = Planck's constant, J s
j = imaginary unit
$\boldsymbol{k}$ = phonon-mode wavenumber, m^{-1}
K = modified Bessel function of the second kind
l = angular momentum quantum number
MFP = mean free path
P_l = spherical harmonic of order l
$Q(x)$ = net heat flux
t = time, s
T = temperature, K
v = group-velocity magnitude
∇ = gradient operator
θ = angle of wavevector with respect to transport axis
κ = thermal conductivity, W/m K
Λ_i = mean free path of phonon mode indexed by "i"
$\tau_i\ (k)$ = lifetime of a mode indexed by i with wavevector magnitude k, s
χ = transformation of "x" coordinate variable, m
ω = angular frequency

Appendix A

Here, we suggest ways of generalizing the one-dimensional analysis and generalize the EFL to three spatial dimensions.

Although we have truncated the spherical harmonic expansion of the distribution function at the $l=2$ order, it is possible to generalize to arbitrary order. Equations (4a)–(4c) are part of a hierarchy of equations, the lth equation of which is, for $l>0$ [22]

$$\frac{l+1}{2l+3}v\frac{\partial g_{l+1}}{\partial x}+\frac{l}{2l+1}v\frac{\partial g_{l-1}}{\partial x}+\frac{g_l}{\tilde{\tau}}=0 \tag{A1}$$

Of course, increasing the number of equations in the hierarchy improves the accuracy of the model. However, this will result in a differential equation of higher order, requiring more boundary conditions than can be deduced from the physics of the problem.

In three dimensions, the equations of the generalized EFL and energy conservation take the following form [23]:

$$\left(1+\tau_i\frac{\partial}{\partial t}\right)^2\boldsymbol{q}_i = -\kappa_i\nabla T+\frac{3}{5}\left(\Lambda_i^{\rm LF}\right)^2\nabla(\nabla\cdot\ \boldsymbol{q}_i) -\frac{1}{5}\left(\Lambda_i^{\rm LF}\right)^2\nabla\times(\nabla\times\ \boldsymbol{q}_i) \tag{A2}$$

$$\nabla\cdot\left(\sum_i\boldsymbol{q}_i\right)=-C_v\frac{\partial T}{\partial t} \tag{A3}$$

where $\boldsymbol{q}_i$ is the low-frequency quasi-ballistic mode indexed by i, $\Lambda_{\rm i}$ is its mean free path, κ_i is the kinetic theory value of the each mode's thermal conductivity, T is the temperature, τ_i is the lifetime of phonons in the ith channel, and C_v is the heat capacity of all phonon modes combined. Although Ramu and Bowers [23] derived this equation starting from the steady-state BTE, extension to time-dependent BTE is simple using Fourier transforms, as illustrated in Sec. 3. The last term of Eq. (A2) is a circulatory term. Inclusion of this term requires knowledge of the tangential heat flux at the surface, which is not available except for white, specular boundaries. The physics of this term has only recently been touched upon [24] and we will not concern ourselves with it in this paper.

Appendix B

Although periodicity in space has been applied to test our expressions against the benchmark of Maznev et al., it is by no means necessary— inverse Fourier transforms yield the requisite differential equations in cases where there are boundaries present and the conditions there need to be considered. Our formulation has the important advantage of yielding families of generalized Fourier laws of various finesse since $A_{\mathrm{EFL}}(\chi\Lambda)$ is the ratio of rational polynomials. To wit, we use this formulation to generate a generalized EFL for a material whose MFP spectrum can be decomposed into a high-frequency (HF) channel and two low-frequency (LF) channels.

To this end, consider a cut-off mean free path $\Lambda_F = \Lambda(k_F)$ such that $\Lambda^2\chi^2 \ll 1$ for all length scales $1/\chi$ of interest and all $\Lambda < \Lambda_F$. For $\Lambda \geq \Lambda_F$, let the differential conductivity be given by

$$\kappa_{\mathrm{diff}}(k) = \Delta\kappa_1\delta(\Lambda - \Lambda_F) + \Delta\kappa_2\delta(\Lambda - \Lambda_2) \tag{B1}$$

where δ is the Dirac delta function.

Substituting Eq. (B1) in Eq. (8) and simplifying, we get

$$\begin{aligned}&\left(1+\frac{3}{5}\Lambda_F^2\chi^2\right)\left(1+\frac{3}{5}\Lambda_2^2\chi^2\right)(Q(\chi)+i\chi T\kappa_F)\\&= \Delta\kappa_1(-i\chi T)\left(1+\frac{3}{5}\Lambda_2^2\chi^2\right)+\Delta\kappa_2(-i\chi T)\left(1+\frac{3}{5}\Lambda_F^2\chi^2\right)\end{aligned} \tag{B2}$$

where using the assumption $\Lambda^2\chi^2 \ll 1$, we have defined

$$\kappa_F = \int_{k=0}^{k_F} \frac{\kappa_{\mathrm{diff}}(k)}{1+\frac{3}{5}\Lambda^2\chi^2}dk \sim \int_{k=0}^{k_F} \kappa_{\mathrm{diff}}(k)dk \tag{B3}$$

We may easily take the inverse Fourier transform of Eq. (B3). Discarding fourth-order derivatives with respect to x, we get

$$\begin{aligned}Q(x) &= \frac{3}{5}\left(\Lambda_2^2+\Lambda_F^2\right)\frac{\partial^2 Q}{\partial x^2} - \kappa_{\mathrm{bulk}}\frac{\partial T}{\partial x}\\&\quad+\frac{3}{5}\left[\Lambda_F^2(\kappa_{\mathrm{bulk}}-\Delta\kappa_1)+\Lambda_2^2(\kappa_{\mathrm{bulk}}-\Delta\kappa_2)\right]\frac{\partial^3 T}{\partial x^3}\end{aligned} \tag{B4}$$

where $\kappa_{\mathrm{bulk}} = \kappa_F + \Delta\kappa_1 + \Delta\kappa_2$ is the bulk thermal conductivity, the sum of all phonon contributions. The generalization of Eq. (B4) to an arbitrary number of quasi-ballistic channels may be performed along similar lines.

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