Nonlocality in microscale heat conduction

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(Dated: December 23, 2014)

Abstract

Thermal transport at short length and time scales inherently constitutes a nonlocal relation between heat flux and temperature gradient. Most quasiballistic studies do not address the nonlocality explicitly, and relatively few works provide direct comparisons between theory and actual measurement results. Here, we present an experimentally validated universal formalism that enables detailed characterisation of the nonlocal effects. Synergy with stochastic frameworks captures the essential transport physics in compact models with easy to understand parameters. We obtain a fully analytical expression for the spatial flux memory in tempered Lévy transport with fractal dimension $\alpha$ and diffusive recovery length $x_R$ and find that nonlocality is physically important over distances $\sqrt{2 - \alpha} x_R$. This is not only relevant to heat conduction in semiconductor alloys but also applies to similar dynamics observed in other disciplines including hydrology and chemistry. Accurate matches in closed form to the effective thermal conductivity measured by transient thermal grating and time domain thermoreflectance experiments yield nonlocality lengths around 0.5 $\mu$m in single crystals and 1–2 $\mu$m in alloys, in good agreement with typical median phonon mean free paths. Our approach can help lead the way to a better understanding and efficient modeling of nondiffusive transport regimes.

PACS numbers: 44.10.+i, 65.40.-b, 63.20.-e, 05.60.-k, 05.40.Fb

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I. INTRODUCTION

Microscale heat conduction in semiconductor materials has received rapidly intensifying interest over the past years [1–13]. These transport regimes not only have immediate practical relevance for electronic devices [14] but also provide invaluable clues from which fundamental properties of the microscopic heat carriers can be reconstructed [8, 15, 16].

In regular diffusive transport, Fourier's famous law relates the heat flux $\vec{q}$ to the local temperature gradient $\vec{\nabla}T$ through the medium’s thermal conductivity $\kappa_0$. In the case of one-dimensional heat flow, we have

$$q(x,t) = -\kappa_0 \frac{\partial T}{\partial x}(x,t) \quad (1)$$

However, at length and/or time scales comparable to phonon mean free paths (MFPs) and/or relaxation times, the Fourier law breaks down, and the transport no longer behaves as predicted by standard diffusive theory. Prime observations of such ‘quasiballistic’ regimes have been achieved through time domain thermoreflectance (TDTR) [1, 3, 16] and transient thermal grating (TTG) [4, 5] experiments. A growing body of work has been undertaken to develop a deeper understanding of the underlying physics [7, 9–13, 15, 16].

Quasiballistic transport inherently causes the heat flux in a given location at a given time to co-depend on the temperature gradient in other places and/or earlier times [17–19]. Put differently, the flux-gradient relation (FGR) is no longer localised, but instead possesses ‘memory’ in space and/or time. Building upon the pioneering work by Mahan and Claro for steep gradients [17], Koh and coworkers recently proposed a nonlocal heat conduction theory [20] to investigate quasiballistic transport observed in TDTR experiments on semiconductor alloys [1]. Although this work enabled validation of the main trends, the theory explicitly relied on a set of simplifying assumptions in an ideal Debye crystal and consequently only applies to ‘weakly quasiballistic’ regimes in the high temperature limit. Moreover, the nonlocal heat equation had to be solved numerically with a finite element scheme that required several thousands of nodes. Despite the severe computational cost, the theory still lacked flexibility for direct comparison with actual measurement results, and therefore is unable to characterise the heat conduction nonlocality in a given test sample.

In this work, we provide a universal nonlocal formalism that can directly determine the shape of the heat flux delocalisation and distance over which it is physically important from
experimental data. Our theory can be applied across the entire ballistic-diffusive spectrum and ambient temperature range, and readily accommodates complex crystal structures with arbitrary phonon dispersions. The essential physics of quasiballistic transport regimes can be captured in just a few parameters through synergy with stochastic frameworks, enabling a fully analytical study of FGR delocalisation in semiconductor alloys. We also obtain closed form expressions for the effective thermal conductivity as a function of grating period and laser modulation frequency that yield good agreement with respective TTG and TDTR measurements. We find associated nonlocality distances $\approx 0.5 \, \mu m$ in single crystal materials and $2–3 \, \mu m$ in alloys, being on the order of typical median phonon MFPs for both cases, as intuitively expected.

II. THEORETICAL FORMALISM

A. Nonlocal framework

We assume a homogeneous, isotropic medium and perform the derivation for one-dimensional heat flow. A 1D framework will suffice for our purposes, as most TTG and TDTR experiments are dominated by in-plane and cross-plane transport respectively. We note that explanation of laser spot size effects [3] does require detailed 3D analysis [12] and will not be attempted here. We seek to extend the conventional Fourier law (1) to nondiffusive heat conduction regimes in the form

$$q(x, t) = -\int_0^t dt' \int_{-\infty}^{\infty} dx' \kappa^*(x - x', t - t') \frac{\partial T}{\partial x}(x', t')$$

Here, $\kappa^*(x', t')$ is a generalised thermal conductivity kernel that embodies the spatial and temporal nonlocality of the heat flux with respect to the temperature gradient. We stress this quantity must not be confused with the ‘effective’ conductivity $\kappa_{\text{eff}}$ that arises in phenomenological interpretations $q(x, t) = -\kappa_{\text{eff}}(x, t) \partial_x T(x, t)$ of nondiffusive transport as is commonplace in virtually all quasiballistic heat experiments [1–6]. Following Fourier ($x \leftrightarrow \xi$) and Laplace ($t \leftrightarrow s$) transformations, the double convolution in (2) simplifies to

$$q(\xi, s) = j\xi \kappa^*(\xi, s) T(\xi, s)$$

3
with \( j \) the complex unit. In a medium with volumetric heat capacity \( C_0 \), conservation of energy requires that \(-\partial_x q = C_0 \partial_t T\) at all places and times, and hence

\[
-j\xi q(\xi, s) = sP(\xi, s) - P(\xi, t = 0)
\]

where we introduced the volumetric energy density \( P = C_0 T \). Both \( T \) and \( P \) express deviations relative to the ambient background. We now consider a planar source at \( x = 0 \) inside the medium that injects 1 J/m\(^2\) at time zero to find the single pulse energy density response \( P(\xi, s) \) from which all dynamic system properties can be derived. Given the initial condition \( P(x, t = 0) = \delta(x) \) we have \( P(\xi, t = 0) = 1 \), so combining (3) and (4) produces

\[
\kappa^*(\xi, s) = \frac{C_0}{\xi^2} \left[ \frac{1}{P(\xi, s)} - s \right]
\]

Our derivation solely relied on energy conservation and basic isotropy, without making any other assumptions about the medium’s crystal configuration or phonon characteristics. The result (5) is therefore universally applicable to all transport regimes across the ballistic-diffusive spectrum in a wide variety of materials.

We can connect the macroscopic kernel \( \kappa^* \) to the underlying microscopic dynamics by solving the 1D time dependent Boltzmann transport equation (BTE) under the relaxation time approximation. This provides a fully analytical expression for \( P(\xi, s) \) in terms of the fundamental phonon properties [15]. Inserting this BTE solution into (5) yields

\[
\kappa^*(\xi, s) = \left( \sum_k C_k \right) \cdot \left( \sum_k \frac{\kappa_k}{(1 + s\tau_k)^2 + \xi^2\Lambda_{||,k}^2} \right) / \left( \sum_k \frac{C_k (1 + s\tau_k)}{(1 + s\tau_k)^2 + \xi^2\Lambda_{||,k}^2} \right)
\]

Here, \( \kappa_k = \Lambda_{||,k} v_{||,k} C_k \) is the thermal conductivity of a phonon mode resolved for wavevector \( \vec{k} \), with mean free path (MFP) \( \Lambda_k = v_k \tau_k \), group velocity \( v_k \), volumetric heat capacity \( C_k \) (unit J/m\(^3\)-K) and relaxation time \( \tau_k \). Subscripts \( || \) indicate \( x \)-projection onto the thermal transport axis. The bulk thermal diffusivity \( D_0 = \kappa_0/C_0 \) is given by \( \sum_k \kappa_k / \sum_k C_k \). The summations running over a discrete wavevector grid in (6) enable seamless analysis of ‘ab initio’ phonon data, but can be easily converted to continuous integrals over wavevector space or phonon frequency for analytical modelling. In particular, for an idealised crystal...
with lattice constant \( a \) and spherically symmetric Brillouin zone (BZ) we have

\[
\kappa_{SBZ}(\xi, s) = C_0 \left( \frac{1}{(2\pi)^2} \int_0^\pi \sin \theta d\theta \int_0^{\pi/a} k^2 dk \Lambda(k) v(k) C(k) \cos^2 \theta \right) \left( \frac{1}{(2\pi)^2} \int_0^\pi \sin \theta d\theta \int_0^{k_{\text{max}}} k^2 dk \frac{C(k) [1 + s \tau(k)]}{[1 + s \tau(k)]^2 + \xi^2 \Lambda^2(k) \cos^2 \theta} \right) \tag{7}
\]

where \( C_0 = \frac{1}{2\pi^2} \int_0^{\pi/a} C(k) k^2 dk \). We note that in integral formulations \( C(k) \) denotes the heat capacity of a single mode (unit J/K and equal to the Boltzmann constant in the high temperature limit) and that summation over phonon branches is implicitly assumed in both (6) and (A4). It can be shown that the nonlocal model developed by Koh et al. \cite{20} formally corresponds to a simple special case of the much more general framework presented here \cite{21}. We also see that when \( \xi \Lambda \ll 1 \) and \( s \tau \ll 1 \), the master expression (6) reduces to \( \kappa^*(\xi, s) = \kappa_0 \), and hence \( \kappa^*(x', t') = \kappa_0 \delta(x')\delta(t') \). Thus, for transport occurring over length and time scales that are sufficiently larger than phonon MFPs and relaxation times, the FGR described by (2) becomes localised again and recovers to Fourier’s law, as appropriate.

### B. Synergy with stochastic frameworks

Stochastic transport models, which describe the governing dynamics in terms of random motion of energy carriers, have been well established in the literature for a wide variety of applications \cite{22,24}. We briefly review the key fundamentals relevant to our discussion of quasiballistic heat conduction. Within the stochastic context, the single pulse response \( \mathcal{P}(x, t) \) expresses the chance to find a randomly wandering energy packet in location \( x \) at time \( t \) after it was released by the source in \( x = 0 \) at \( t = 0 \). In the case of 1D flight processes, the packet moves through consecutive jumps \( x(t + \vartheta) = x(t) + \zeta \) where the jump length \( \zeta \) and wait time \( \vartheta \) are randomly chosen from stochastically independent distributions \( \phi(\zeta) \) and \( \varphi(\vartheta) \). The Montroll-Weiss equation \cite{24} provides a closed form solution in Fourier-Laplace domain for the single pulse response induced by the flight process:

\[
\mathcal{P}(\xi, s) = \frac{1 - \varphi(s)}{s[1 - \phi(\xi)\varphi(s)]} = \frac{\Psi(s)}{s[\Psi(s) + \psi(\xi)]} \tag{8}
\]
where we introduced $\psi(\xi) = 1 - \varphi(\xi)$ and $\Psi(s) = 1/\varphi(s) - 1$ for notation convenience. The associated generalised conductivity kernel given by (5) then reads

$$\kappa^*(\xi, s) = C_0 \cdot \frac{\psi(\xi)/\xi^2}{\Psi(s)/s}$$  \hspace{1cm} (9)

In ‘Poissonian’ flight processes $\Psi(s) = s\vartheta_0$ (the wait time distribution is exponential with average $\vartheta_0$), the single pulse response (8) and generalised conductivity (9) simplify to

Poissonian: $\mathcal{P}(\xi, s) = \frac{1}{s + \psi(\xi)/\vartheta_0}$, \hspace{1cm} $\kappa^*(\xi) = C_0 \frac{\psi(\xi)}{\vartheta_0 \xi^2}$ \hspace{1cm} (10)

The $s$ dependence has vanished from $\kappa^*$: the FGR in Poissonian transport has no temporal memory. Since the master equation (6) becomes equally insensitive to $s$ whenever $s\tau \ll 1$, ‘weakly quasiballistic’ transport \[11\] is inherently Poissonian and only delocalised in space. Relaxation times of the dominant heat carriers in semiconductors typically span the sub-ps to 2–3 ns range \[15\], with the longest living modes having the smallest density of states. We can therefore reasonably assume that the Poissonian regime extends up to $s\tau_{\text{max}} \leq 1$, or roughly $f \leq 50$ MHz, and thus encompasses the typical operational range of TTG and TDTR experiments \[11, 20\].

Several subtypes of Poissonian flight processes deserve specific mentioning. The first consists of regular Fourier diffusion, whose single pulse response $\mathcal{P}(\xi, s) = 1/(s + D_0\xi^2)$ corresponds to $\psi(\xi) = L_0^2\xi^2$ with $L_0^2/\vartheta_0 = D_0$. From (10) we have $\kappa^*(\xi, s) \equiv \kappa_0$ so the FGR correctly recovers again to Fourier’s law. Second, $\psi(\xi) = L_0|\xi|$ describes a ballistic regime with Cauchy-Lorentz distributions $\mathcal{P}(\xi, s) = 1/(s + v_0|\xi|)$ and $\kappa^*(\xi) = C_0 v_0/|\xi|$ where $v_0 = L_0/\vartheta_0$ is the average propagation velocity. Finally, intermediate cases $\psi(\xi) = L_0^\alpha |\xi|^\alpha$ with fractional exponents $1 < \alpha < 2$ correspond to Lévy superdiffusion with fractal dimension $\alpha$ and fractional diffusivity $D_\alpha = L_0^\alpha/\vartheta_0$ (unit m$^\alpha$/s). The single pulse response is governed by alpha-stable distributions which have characteristic ‘heavy tails’ $\mathcal{P}(x \gg [D_\alpha t]^{1/\alpha}, t) \sim |x|^{-(\alpha + 1)}$. Now $\kappa^*(\xi) = C_0 D_\alpha/|\xi|^{2 - \alpha}$ so the spatial heat flux memory decays algebraically with distance: $\kappa^*(x') \sim |x'|^{-(\alpha - 1)}$. The characteristic blueprints for the three discussed cases are summarised in Table I.

III. RESULTS

Using ab initio phonon populations courtesy of J. Carrete and N. Mingo \[15\] as input to Eq. (6), we evaluate the theoretical FGR memory kernel in Si, InGaAs and SiGe.
TABLE I. Spatial heat flux memory kernel $\kappa^*(\xi)$ for Poissonian transport in isotropic medium with bulk conductivity $\kappa_0$ and volumetric heat capacity $C_0$.

| Transport regime          | Governing Parameter(s) $\kappa^*(\xi)$ [W/m-K] |
|---------------------------|--------------------------------------------------|
| Fourier diffusion         | bulk diffusivity $D_0$                          |
|                            | $C_0D_0 = \kappa_0$                            |
| Lévy superdiffusion       | fractal dimension $1 < \alpha < 2$               |
|                            | fractional diffusivity $D_\alpha$               |
|                            | $C_0D_\alpha/|\xi|^{2-\alpha}$                   |
| Ballistic (Cauchy-Lorentz)| avg. propagation velocity $v_0$                  |
|                            | $C_0v_0/|\xi|$                                    |

normalised magnitude of generalised conductivity $|\kappa^*(\xi, j2\pi f)| / \kappa_0$

phase of generalised conductivity $\angle[\kappa^*(\xi, j2\pi f)]$

FIG. 1. Space and time heat flux memory kernel $\kappa^*(\xi, s = j2\pi f)$ evaluated from Eq. (6) with ab initio phonon populations for Si, InGaAs and SiGe. The purely diffusive and Poissonian quasiballistic regimes can be easily identified in the magnitude plots.

Full transient solutions $\kappa^*(\xi, s)$ (Fig. 1) become virtually independent of temporal frequency below the 40–80 MHz range, in good agreement with the 50 MHz Poissonian threshold anticipated earlier. From the Poissonian limit $\kappa^*(\xi, s \to 0)$ (Fig. 2) we can easily identify distinct regimes based on the characteristic signatures from Table I. In Si we observe a fairly smooth, continuous transition from ballistic transport at short length scales to regular diffu-
FIG. 2. Spatial heat flux memory kernel $\kappa^*(\xi)$ for Poissonian transport, i.e. transients that are slow compared to phonon relaxation times, calculated from ab initio phonon populations. Distinct regimes are easily identified using Table I. The emergence of fractal Lévy dynamics with $\alpha \simeq 5/3$ in alloy materials is clearly visible.

InGaAs and SiGe, by contrast, exhibit an additional intermediate regime where $\kappa^* \sim \xi^{-1/3}$, indicating Lévy transport with fractal dimension $\alpha \simeq 5/3$. The emergence of Lévy dynamics in semiconductor alloys has only been pointed out very recently [15, 16] and physically originates in their large phonon scattering exponent. Starting from (A4), it is possible to show that a simplified Debye crystal in Poissonian regime with scattering relation $\Lambda(k) \sim k^{-n}$ ($n > 3$) generates Lévy transport with $\alpha = 1 + 3/n$ [21].

We can investigate the FGR delocalisation in semiconductor alloys more closely with a simple model for the Lévy-Fourier transition. Recalling the functional forms in Table I for Poissonian flight processes, we set:

$$D^*(\xi) = \frac{\kappa^*(\xi)}{C_0} = \frac{D_0}{(1 + x_{LF}^2 \xi^2)^{1-\alpha/2}} \quad (1 < \alpha < 2)$$  \hspace{1cm} (11)

For $x_{LF} \xi \gg 1$ this corresponds to Lévy transport with fractal dimension $\alpha$ and fractional diffusivity $D_\alpha = D_0/x_{LF}^{2-\alpha}$ while $x_{LF} \xi \ll 1$ yields regular diffusion with bulk diffusivity $D_0$. The parameter $x_{LF}$ thus acts as a characteristic length scale for diffusive recovery, and we expect it to be on the order of median MFPs (a few microns in typical alloys). Note that (11) describes a generic transition from Lévy transport to regular diffusion without making any assumptions about the nature of the microscopic carriers. The analysis that follows, therefore, is not limited to heat conduction in alloys but can also provide valuable insight to other disciplines where similar dynamics have been observed such as sediment transport in rivers [25, 26] and solute chemical reactions in flows [27, 28].
A key advantage of (11) is that it can be transformed analytically to real space domain:

\[
D^*(x') = \frac{2^\nu D_0}{\sqrt{\pi} \Gamma(1/2 - \nu)} \frac{K_\nu(|x'/x_{\text{LF}}|)}{|x'/x_{\text{LF}}|^{\nu}}, \quad \nu = \frac{\alpha - 1}{2}
\]

where \(K\) is the modified Bessel function of the second kind. The inner core of the kernel exhibits pure Lévy behaviour \(D^*(|x'| \ll x_{\text{LF}}) \sim |x'/x_{\text{LF}}|^{-(\alpha-1)}\), while the tails decay much more rapidly as \(|x'/x_{\text{LF}}|^{-\alpha/2} \exp(-|x'/x_{\text{LF}}|)\). Given that \(D^*(\xi = 0) = D_0\), the total kernel content \(\int_{-\infty}^{\infty} D^*(x')dx'\) always equals the bulk diffusivity. The tempered Lévy parameters regulate how this fixed memory budget gets spatially distributed: \(x_{\text{LF}}\) sets the overall characteristic length scale, while \(\alpha\) governs the shape details. For example, smaller (more superdiffusive) \(\alpha\) values produce kernels that are less sharply concentrated near the origin in favour of stronger tails (Fig. 3), consistent with the notion that delocalisation should become more prominent as transport dynamics deviate further from regular diffusion.

FIG. 3. Flux memory kernel in real space domain for tempered Lévy transport as evaluated from analytical solution (12). \(\alpha\) denotes the fractal dimension of the Lévy regime while \(x_{\text{LF}}\) sets the length scale over which the transport recovers to regular diffusion. The characteristic ‘width’ of the kernel, which acts as a measure for the distance over which nonlocal transport effects are physically important, equals \(\sqrt{2 - \alpha} x_{\text{LF}}\) and is indicated by the arrow marks.
We can easily quantify the associated spatial extent over which nonlocal effects are physically important by considering $D^*(x')/D_0$. This quantity acts like a properly normalised density function for which we analytically obtain a standard deviation $\sqrt{\langle X'^2 \rangle} = \sqrt{2 - \alpha} \ x_{LF}$. Notice that $\langle X'^2 \rangle \to 0$ in the diffusive limit $\alpha \to 2$, signaling a collapse of the memory kernel into a single Dirac peak as appropriate.

IV. APPLICATIONS

A. Transient thermal grating experiments

TTG employs the interference pattern of two laser beams to impose a heat source that is periodic in space [5]. This produces a sinusoidal temperature profile with grating period $\lambda$ at the semiconductor surface that then equilibrates predominantly through 1D in-plane transport. TTG thus essentially probes the medium’s Fourier domain response at a single spatial frequency $\xi_\lambda = 2\pi/\lambda$. In diffusive regime, the peak-to-valley temperature contrast of the grating pattern decays exponentially with time constant $\Theta_0 = 1/D_0 \xi_\lambda^2 = \lambda^2/4\pi^2D_0$ [5].

Grating periods that overlap with phonon MFPs induce nondiffusive transport. Most experiments probe a regime where the BTE solution still decays exponentially, but slower than predicted by diffusive theory [11]. Identifying the time constant $\Theta$ of an exponential fit to the measured quasiballistic transient with the diffusive expression $\Theta_0$ provides the ‘effective’ thermal conductivity $\kappa_{\text{eff}}(\lambda) = C_0/\Theta \xi_\lambda^2$ [5].

The exponential decay of the measured responses is a direct manifestation of Poissonian transport dynamics. Indeed, at a given spatial frequency $\xi_\lambda$ the generic pulse response of a Poissonian flight process (10) decays exponentially in time with time constant $\Theta = \vartheta_0/\psi(\xi_\lambda)$. Moreover, from (10) we obtain $\kappa^*(\xi_\lambda) = C_0\psi(\xi_\lambda)/\vartheta_0\xi_\lambda^2 = C_0/\Theta \xi_\lambda^2$, which is identical to $\kappa_{\text{eff}}(\lambda)$ derived earlier. The effective conductivity inferred at grating period $\lambda$ thus formally corresponds to a direct sampling of the heat flux memory $\kappa^*(\xi)$ at spatial frequency $\xi_\lambda$. This renders TTG experiments highly suitable for direct exploration of FGR delocalisation.

Using these insights, we apply our framework to measurements performed by Johnson and coworkers on bulk GaAs and Si membranes [4]. To simplify the analysis, we model the transport in these single crystal materials with the functional form

$$\kappa^*(\xi) = \frac{\kappa_0}{(1 + x_{\text{FPS}}m)^{1/m}}$$

(13)
It is easy to verify this corresponds to a transition from ballistic Cauchy transport to regular Fourier diffusion over characteristic length scale $x_{CF}$. The exponent $m$ regulates the sharpness of the regime cross-over. Physically, $x_{CF}$ holds information about the core MFPs while $m$ can be closely associated with the shape and extent of the upper tail of the cumulative conductivity spectrum $\kappa_\Sigma(\Lambda)$ \cite{21}. The special case $m = 2$ corresponds to the limit $\alpha \to 1$ of the tempered Lévy framework \cite{11} considered earlier, where $\kappa^*(x') = (\kappa_0/\pi x_{CF})K_0(|x/x_{CF}|)$. Evaluating $\kappa^*(\xi = 2\pi/\lambda)$ for the general case yields

$$\kappa_{eff}(\lambda) = \frac{\kappa_0}{[1 + (2\pi x_{CF}/\lambda)^m]^{1/m}}$$  \hspace{1cm} (14)

Since this expression results from a Poissonian framework, its accuracy should be expected to diminish for very small grating periods as then the characteristic time scale $\Theta \sim \lambda^2$ becomes comparable with phonon relaxation times. This brings temporal flux memory into play, and the non-Poissonian transport corresponds to the ‘strongly quasiballistic’ regime in which the BTE solution can no longer be fitted by a simple exponential decay \cite{11}. At room temperature we estimate crossover in typical semiconductors around $\lambda \simeq 1 \mu m$. The experiments with $\lambda \geq 2 \mu m$ reported in Ref. \cite{4} should thus operate within the Poissonian regime, and as a result our simple model indeed provides an accurate fit (Fig. 4).

![Graph showing thermal transient grating measurement results](image)

**FIG. 4.** Thermal transient grating measurement results agree well with our analytical expression \cite{14}. This provides the ballistic-diffusive recovery length scale $x_{CF}$ over which heat conduction nonlocality is physically important. A uniform scaling factor of 0.62 was applied to the membrane model results to account for thin film suppression of the bulk conductivity.

Identified $x_{CF}$ values amount to 600 nm for bulk GaAs and 400 nm for the Si membrane, in good agreement with typical median MFPs \cite{29}. The smaller $x_{CF}$ and larger $m$ of the Si membrane is testament to the compression of the upper regions of the MFP spectrum due
to thin film boundary scattering [30]. It is noteworthy that our expression (14) matches the experimental data considerably better than other theories previously reported by the original experimentalists [4, 5]. Good analytical forms for $\kappa_{\text{eff}}$ can be quite valuable in practice, as they would facilitate the ill-posed MFP reconstruction problem [8]. Several studies [7, 9, 11] carried out detailed analyses of TTG quasiballistic physics but, interestingly, none of them provided a direct comparison between theory and experiments. One possible reason is that the strong reliance on Callaway scattering models and Debye-type crystals in these works proves too inflexible or complicated to analyse actual measurement data. The viewpoint adopted here, by contrast, captures the essential dynamics in stochastic models with a minimum of parameters that provide direct physical insight.

B. Time domain thermoreflectance experiments

TDTR techniques use laser pulse trains that are modulated at temporal frequency $f_{\text{mod}}$ as heat source [1, 31]. The thermal dynamics of the semiconductor can only be studied indirectly since the method requires a thin metal film to be deposited onto the sample surface as measurement transducer. While this and other factors complicate the processing of raw measurement data [31], the measured transient is dominated by 1D cross-plane transport and primarily sensitive to the frequency domain single pulse response $P(x = 0, s = j2\pi f_{\text{mod}})$ of the semiconductor surface [20, 31]. We will therefore ignore the presence of the transducer and again assume an infinite medium with heat source at $x = 0$. This simplification will prove sufficient for our main purpose here, which is to use our nonlocal framework to explain the experimental frequency dependence $\kappa_{\text{eff}}(f_{\text{mod}})$ in semiconductor alloys [1].

In diffusive regime, a modulated heat source induces an exponential temperature gradient inside the medium:

$$\frac{\partial T}{\partial x}(x) \sim \exp(-|x|/\ell)$$

(15)

The decay length $\ell = \sqrt{D_0/\pi f_{\text{mod}}}$ ranges from $\approx 1\,\mu\text{m}$ at 1 MHz to $\approx 250\,\text{nm}$ at 20 MHz in typical semiconductor alloys. The considerable overlap with phonon MFPs induces quasibal-
listic transport that is conventionally analysed with ‘modified Fourier’ theory. This means a purely diffusive model is still used, but the bulk conductivity $\kappa_0$ is phenomenologically adjusted with modulation frequency to fit the data as best as possible [1]. The analysis therefore still assumes an exponential gradient, but with an ‘effective’ decay length $\ell_{\text{eff}}$. We
can evaluate the apparent conductivity \( \kappa_{\text{eff}}(x) = -q(x)/\partial_x T(x) \) that would be inferred at depth \( x \) by applying the convolution relation \[2\] to the functional form \[15\]:

\[
\kappa_{\text{eff}}(x) = \exp(|x|/\ell_{\text{eff}}) \int_{-\infty}^{\infty} \kappa^*(x - x') \exp(-|x'|/\ell_{\text{eff}}) dx'
\]

We could safely ignore the time component of the convolution since the experimental bandwidth operates within the Poissonian regime as determined earlier. After expressing the spatial memory kernel in terms of its Fourier image

\[
\kappa^*(x - x') = \frac{1}{\pi} \int_0^\infty \kappa^*[\xi(x - x')] d\xi
\]

the integration over \( x' \) in \[16\] can be carried out analytically, and we obtain

\[
\kappa_{\text{eff}}(x) = \frac{2\ell_{\text{eff}}}{\pi} \exp(|x|/\ell_{\text{eff}}) \int_0^\infty \frac{\kappa^*(\xi) \cos(\xi x) d\xi}{1 + \xi^2 \ell_{\text{eff}}^2}
\]

Notice that, contrary to TTG, the effective conductivity inferred by TDTR is determined by a broad spatial frequency sampling of the \( \kappa^*(\xi) \) kernel. Expressing that \( \ell_{\text{eff}} = \sqrt{\kappa_{\text{eff}}/(\pi C_0 f_{\text{mod}})} \) within the modified Fourier framework leads to an integral equation for the desired \( \kappa_{\text{eff}} \):

\[
\int_0^\infty \frac{\kappa^*(\xi) \cos(\xi x) d\xi}{\pi C_0 f_{\text{mod}} + \kappa_{\text{eff}}(x) \xi^2} - \sqrt{\frac{\pi \kappa_{\text{eff}}(x)}{4 C_0 f_{\text{mod}}} \exp \left( -\sqrt{\frac{\pi C_0 f_{\text{mod}} x^2}{\kappa_{\text{eff}}(x)}} \right) = 0
\]

We will model the thermal transport in the alloy with the tempered Lévy framework \[11\] discussed earlier. In pure Lévy regime \( \kappa^*(\xi) \simeq \kappa_0/|x_{\text{LF}}\xi|^{2-\alpha} \) the effective conductivity at the surface \( x = 0 \) as probed by the measurement can be evaluated analytically:

\[
\kappa_{\text{eff}}(x = 0) = \frac{\kappa_0^{2/\alpha} (\pi C_0 x_{\text{LF}}^2 f_{\text{mod}})^{-(2/\alpha - 1)}}{\cos^{2/\alpha} \left( \frac{\pi}{2} \right)}
\]

This predicts an algebraic dependence on modulation frequency, which matches well with our experimental results for InGaAs and SiGe as shown in Fig. 5.

The exponents of the power law fits indicate fractal dimensions \( \alpha = 1.67 \) for InGaAs and \( \alpha = 1.71 \) for SiGe, in near perfect agreement with values determined through full 3D analysis of raw experimental data \[16\] and with the ab initio results discussed earlier in Fig. 2. With \( \alpha \) determined, the prefactor of the power law fit provides the Lévy-Fourier transition
FIG. 5. The frequency dependence of effective conductivity in time domain thermoreflectance experiments agrees well with our analytical expression [20]. A simple power law fit provides the fractal dimension $\alpha$ of the Lévy heat conduction inside the alloy and the diffusive recovery length $x_{LF}$. Nonlocality is physically important over distances $\sqrt{2 - \alpha} x_{LF} \simeq x_{LF}/2$.

length once bulk thermal properties are specified. We obtain $1.6 \, \mu m \leq x_{LF} \leq 3.8 \, \mu m$ in InGaAs ($\kappa_0 = 6 - 8 \, \text{W/m-K}$) and $1.75 \, \mu m \leq x_{LF} \leq 4.7 \, \mu m$ in SiGe ($\kappa_0 = 4.5 - 6 \, \text{W/m-K}$). The obtained $x_{LF}$ values are several times larger than $\ell_{eff}$ over most of the measurement bandwidth, showing that a pure Lévy approximation was indeed justified.

We also take a look at how the effective conductivity evolves if one could probe the transport at different depths inside the medium. Now [19] must be solved numerically, but this is easily achieved by determining the root of the left hand side functional with a simple bisection method in which the integral is evaluated numerically by adaptive quadrature. Figure 6 shows the results for a generic semiconductor alloy with $\alpha = 5/3$, $x_{LF} = 2 \, \mu m$, $\kappa_0 = 6 \, \text{W/m-K}$ and $C_0 = 1.5 \, \text{MJ/m}^3\text{-K}$.

Just a micron away from the heat source, the effective conductivity progressively exceeds the bulk value as the modulation frequency increases. This ‘enhanced’ transport emerges due to the heavy tails of the Lévy dynamics, which carry some energy further away from the heat source than is possible by regular diffusion. The situation inside the medium is dramatically different from that observed at the surface, where the transport is progressively suppressed. This illustrates that careful attention to experimental geometry is essential when interpreting nondiffusive transport, and provides incentives for the development of new measurement techniques to study the transport underneath the surface.
FIG. 6. Effective conductivity normalised to the bulk value that would be inferred from time domain thermoreflectance experiments if one could probe the sample response at various depths. Results are obtained by numerical solution of integral equation (19) for a generic alloy with bulk thermal parameters $\kappa_0 = 6 \text{ W/m-K}$, $C_0 = 1.5 \text{ MJ/m}^2\text{-K}$ governed by tempered Lévy transport (11) with fractal dimension $\alpha = 5/3$ and diffusive recovery length $x_{LF} = 2 \mu\text{m}$.

Moreover, our results indicate that modified Fourier theory is in effect self-contradictory and therefore poorly suited to describe the quasiballistic dynamics that the TDTR experiments try to characterise, as we can see as follows. To analyse measured responses, modified Fourier theory adjusts the bulk conductivity of the medium. It thus assumes by construction that at a given modulation frequency the thermal transport is suppressed by the same amount everywhere in the medium. As Fig. 6 illustrates, this is not at all the case: the effective conductivity a few microns inside the medium can be an order of magnitude larger than the value perceived at the top surface. These issues can be overcome by using a tempered Lévy framework [16] to interpret the experiments. This produces a superior fit to the raw measurement data with less parameters, and also enables an improved characterisation of the intrinsic thermal resistance of the transducer/semiconductor interface [32].

V. CONCLUSIONS

In summary, we have presented a nonlocal framework for microscale heat conduction in semiconductor materials. Contrary to previous work, our formalism can be easily compared to actual measurement data and provides direct information about the nature and spatial extent of the heat conduction nonlocality. The theory is also more general as it is applicable...
across the entire ballistic-diffusive spectrum and accommodates arbitrary phonon dispersions. Synergy with stochastic theory reduces numerical computation costs to a minimum and captures the fundamental transport physics into simple models with just a few easy to understand parameters. This yielded a fully analytical solution for the spatial flux memory in tempered Lévy transport that can be readily applied to other disciplines. We also obtained closed form expressions for the effective thermal conductivity inferred by transient thermal grating and time domain thermoreflectance that are in good agreement with experiments. Our results offer interesting potential for improved compact thermal modeling and a general framework to study nonlocal memory effects in a wide range of transport applications.

ACKNOWLEDGEMENTS

A.S. acknowledges financial support from Purdue University start up funds.

[1] Y.K. Koh and D.G. Cahill, Phys. Rev. B 76, 075207 (2007).
[2] M.E. Siemens, Q. Li, R. Yang, K.A. Nelson, E.H. Anderson, M.M. Murnane, and H. C. Kapteyn, Nature Mater. 9, 26 (2010).
[3] A.J. Minnich, J.A. Johnson, A.J. Schmidt, K. Esfarjani, K. M.S. Dresselhaus, and G. Chen, Phys. Rev. Lett. 107, 095901 (2011).
[4] J.A. Johnson, A.A. Maznev, J.K. Eliason, A. Minnich, K. Collins, G. Chen, J. Cuffe, T. Kehoe, C.M. Sotomayor Torres, and K.A. Nelson, in MRS Proceedings Vol. 1347, Spring Meeting – Symposium BB (2011).
[5] J.A. Johnson, A.A. Maznev, J. Cuffe, J.K. Eliason, A.J. Minnich, T. Kehoe, C.M. Sotomayor Torres, G. Chen, and K.A. Nelson, Phys. Rev. Lett. 110, 025901 (2013).
[6] K.T. Regner, D.P. Sellan, Z. Su, C.H. Amon, A.J.H. McGaughey, and J.A. Malen, Nat. Commun. 4, 1640 (2013).
[7] A.A. Maznev, J.A. Johnson, and K.A. Nelson, Phys. Rev. B 84, 195206 (2011).
[8] A.J. Minnich, Phys. Rev. Lett. 109, 205901 (2012).
[9] K.C. Collins, A.A. Maznev, Z. Tian, K. Esfarjani, K.A. Nelson, and G. Chen, J. Appl. Phys 114, 104302 (2013).
[10] R.B. Wilson, J.P. Feser, G.T. Hohensee, and D.G. Cahill, Phys. Rev. B 88, 144305 (2013).
[11] C. Hua and A.J. Minnich, Phys. Rev. B 89, 094302 (2014).
[12] D. Ding, X. Chen, and A.J. Minnich, Appl. Phys. Lett. 104, 143104 (2014).
[13] R.B. Wilson and D.G. Cahill, Nat. Commun. 5, 5075 (2014).
[14] E. Pop, S. Sinha, and K.E. Goodson, Proc. IEEE 94, 1587 (2006).
[15] B. Vermeersch, J. Carrete, N. Mingo, and A. Shakouri, Under review with Phys. Rev. B. Preprint available online at arXiv:1406.7341 (2014).
[16] B. Vermeersch, A.M.S. Mohammed, G. Pernot, Y.R. Koh, and A. Shakouri, Under review with Phys. Rev. B. Preprint available online at arXiv:1406.7342 (2014).
[17] G.D. Mahan and F. Claro, Phys. Rev. B 38, 1963 (1988).
[18] S.L. Sobolev, Phys. Rev. E 50, 3255 (1994).
[19] F.X. Alvarez and D. Jou, Appl. Phys. Lett. 90, 083109 (2007).
[20] Y.K. Koh, D.G. Cahill, and B. Sun, Phys. Rev. B 90, 205412 (2014).
[21] See Appendix at end of document for detailed derivation.
[22] R. Metzler and J. Klafter, Phys. Rep. 339, 1 (2000).
[23] R. Metzler and J. Klafter, J. Phys. A 37, R161 (2004).
[24] J. Klafter, A. Blumen, and M.F. Shlesinger, Phys. Rev. A 35, 3081 (1987).
[25] Y. Zhang and M.M. Meerschaert, Water Resourc. Res. 47, W08601 (2011).
[26] Y. Zhang, M.M. Meerschaert, and A.I. Packman, Geophys. Res. Lett. 39, L20404 (2012).
[27] D. Brockmann and L. Hufnagel, Phys. Rev. Lett. 98, 178301 (2007).
[28] D. Bolster, D.A. Benson, T. Le Borgne, and M. Dentz, Phys. Rev. E 82, 021119 (2010).
[29] T. Luo and G. Chen, Phys. Chem. Chem. Phys. 15, 3389 (2013).
[30] W. Liu, K. Etessam-Yazdani, R. Hussin, and M. Asheghi, IEEE Trans. Electron. Dev. 53, 1868 (2006).
[31] D.G. Cahill, Rev. Sci. Instrum. 75, 5119 (2004).
[32] B. Vermeersch, A.M.S. Mohammed, G. Pernot, Y.R. Koh, and A. Shakouri, Phys. Rev. B 90, 014306 (2014).
Appendix A: Comparison with Koh’s nonlocal theory

It is instructive to put the nonlocal model developed by Koh et al. (Ref. 20 in the main manuscript) in perspective with the framework we have developed here. The nonlocal theory in Ref. 20 was constructed under the following explicit assumptions:

1. Measured transients are slow compared to phonon relaxation times \( s \tau \ll 1 \)

2. The medium is an ideal Debye crystal (each phonon mode propagates at the sound velocity \( v_S \)) with spherical Brillouin zone

3. The medium operates under its high temperature regime (all phonon energies obey \( \hbar \omega \ll k_B T_0 \) and consequently \( C(k) \approx k_B \) where \( k_B \) is the Boltzmann constant and \( T_0 \) the absolute ambient temperature)

Calculations were performed in a semi-infinite geometry, which required special care to ‘reflect’ back travelling phonons reaching the top surface back into the medium. This complication can be avoided by instead considering a symmetric configuration of a fully infinite medium with heat source at \( x = 0 \). The flux-gradient relation derived in Ref. 20 then reads

\[
q_{\text{Koh}}(x,t) = -\frac{k_B v_S}{8\pi^2} \int_{-\infty}^{\infty} \int_{0}^{\pi} \sin \theta \mid \cos \theta \mid d\theta \int_{0}^{k_{\text{max}}} k^2 dk \exp \left( -\frac{|x - x'|}{\Lambda(k) \cos \theta} \right) \quad \text{(A1)}
\]

This is compatible with the convolution relation postulated in our Eq. (2) in the main manuscript, with generalised conductivity kernel

\[
\kappa_{\text{Koh}}^{\ast}(x',t') = \frac{k_B v_S \delta(t')}{8\pi^2} \int_{0}^{\pi} \sin \theta \mid \cos \theta \mid d\theta \int_{0}^{k_{\text{max}}} k^2 dk \exp \left( -\frac{x'}{\Lambda(k) \cos \theta} \right) \quad \text{(A2)}
\]

\[
\Rightarrow \kappa_{\text{Koh}}^{\ast}(\xi) = \frac{k_B v_S}{(2\pi)^2} \int_{0}^{\pi} \sin \theta \mid d\theta \int_{0}^{k_{\text{max}}} k^2 dk \frac{\Lambda(k) \cos^2 \theta}{1 + \xi^2 \Lambda^2(k) \cos^2 \theta} \quad \text{(A3)}
\]

Note that the end result is independent of \( s \) since \( (A1) \) is only delocalised in space.
We now recall Eq. (7) from the main manuscript, which evaluates our universal convolution kernel for a crystal with spherical Brillouin zone,

\[
\kappa^*_{\text{SBZ}}(\xi, s) = C_0 \left( \frac{1}{(2\pi)^2} \int_0^\pi \sin \theta \, d\theta \int_0^{k_{\text{max}}} k^2 dk \frac{\Lambda(k) \, v(k) \, C(k) \cos^2 \theta}{[1 + s\tau(k)]^2 + \xi^2 \Lambda^2(k) \cos^2 \theta} \right) \right)
\]

and apply the same set of assumptions \( s\tau(k) \ll 1, v(k) \equiv v_S, C(k) \approx k_B \) that were adopted by Ref. 20. With \( C(k) \) being constant, the denominator of \( \kappa^*_{\text{SBZ}} \) is dominated by phonon modes with large density of states, i.e. those towards the outer regions of the BZ. These modes have MFPs that are significantly smaller than typical length scales of experimentally induced thermal gradients, and we can safely assume \( \xi\Lambda(k) \ll 1 \) for this part of the calculation. Consequently the denominator simply tends to the bulk volumetric heat capacity \( C_0 = (1/2\pi^2) \int_0^{k_{\text{max}}} C(k) k^2 dk \), and (A4) reduces to

\[
\kappa^*_{\text{SBZ}}(\xi) \approx \frac{k_B v_S}{(2\pi)^2} \int_0^\pi \sin \theta \, d\theta \int_0^{k_{\text{max}}} k^2 dk \frac{\Lambda(k) \cos^2 \theta}{1 + \xi^2 \Lambda^2(k) \cos^2 \theta} \quad (A5)
\]

which is identical to (A3). The model developed independently by Koh and coworkers in Ref. 20 based on the Mahan-Claro theory thus formally corresponds to a simple special case of the formalism we presented here. Contrary to previous work, our framework is applicable to fully ballistic \( (s\tau \geq 1) \) and low temperature regimes, and can readily account for complex Brillouin zone geometries with arbitrary phonon dispersions.

**Appendix B: Emergence of Lévy dynamics in alloy materials**

Ab initio results for InGaAs and SiGe presented in Fig. 2 of the main manuscript clearly exhibit Lévy features \( \kappa^*(\xi) \sim |\xi|^{-(2-\alpha)} \). The physical origin of this behaviour can be understood based on the generalised conductivity (A5) of a Debye crystal with spherical BZ in Poissonian regime derived above. To simplify the analysis, we replace \( \cos^2 \theta \) (where \( \theta \) sweeps from 0 to \( \pi \)) in the \( k \) integral by its average value 1/2 and operate under a full continuum limit (lattice constant \( a \to 0 \Rightarrow k_{\text{max}} \to \infty \)). This yields

\[
\kappa^*(\xi) \sim \int_0^\infty \frac{k^2 \Lambda(k) \, dk}{2 + \xi^2 \Lambda^2(k)} \quad (B1)
\]
We further assume a single dominant phonon scattering mechanism $\tau(\omega) \sim \omega^{-n}$ ($n$ not necessarily integer), which under a Debye approximation $v(k) \equiv v_S$ corresponds to $\Lambda(k) \sim k^{-n}$. Now (B1) can be evaluated fully analytically for $n > 3$, producing

$$\Lambda(k) \sim k^{-n} : \quad \kappa^*(\xi) \sim |\xi|^{-(1-3/n)} \quad \Rightarrow \quad \alpha = 1 + \frac{3}{n} \quad (n > 3) \quad (B2)$$

This shows the natural emergence of Lévy dynamics, and additionally establishes a direct connection between the macroscopic fractal dimension $\alpha$ and the microscopic phonon scattering exponent $n$. Ideal Rayleigh behaviour in alloys corresponds to $n = 4$ and would induce $\alpha = 7/4$. Experimental values $\alpha \simeq 5/3$ determined by TDTR analysis [16] indicate actual scattering exponents $n \simeq 4.5$, in good agreement with ab initio scattering rates [15]. Notice that the simple model result (B2) maintains pure Lévy transport indefinitely (at all length scales). This is because a purely algebraic scattering relation produces arbitrarily large MFPs near the BZ center $\Lambda(k \rightarrow 0) \rightarrow \infty$ while the continuum limit allows arbitrarily small MFPs $\Lambda(k \rightarrow \infty) \rightarrow 0$. In reality, the MFP spectrum is physically bounded both above and below, causing the Lévy transport to transition to diffusive and ballistic dynamics at respectively long and short length scales as observed in Fig. 2 in the main manuscript.

**Appendix C: Physical meaning of transient thermal grating model parameters**

In the main manuscript, we used the following expression to describe the effective conductivity of single crystal materials inferred by TTG experiments at grating period $\lambda$:

$$\kappa_{\text{eff}}(\lambda) = \frac{\kappa_0}{[1 + (2\pi x_{\text{CF}}/\lambda)^m]^{1/m}} \quad (C1)$$

The figure below shows this functionality for various $m$ values versus normalised grating period $\lambda/x_{\text{CF}}$. It is clear that $x_{\text{CF}}$ sets the overall length scale over which strong quasiballistic effects are induced while $m$ regulates the sharpness of the initial conductivity suppression at large grating periods. Over the considered range $1 \leq m \leq 3$, the grating period where $95\%$ of the bulk conductivity $\kappa_{\text{eff}}(\lambda_{95})/\kappa_0 = 0.95$ is observed spans an order of magnitude ($\lambda_{95} \simeq 119 x_{\text{CF}}$ for $m = 1$ and $\simeq 11 x_{\text{CF}}$ for $m = 3$). Conversely, the midway point where the conductivity suppression is $50\%$ only varies by a factor of 2 ($3.3 x_{\text{CF}} \leq \lambda_{50} \leq 6.3 x_{\text{CF}}$).
FIG. 7. Normalised effective conductivity $\kappa_{\text{eff}}(\lambda)/\kappa_0$ as described by Eq. (C1).

The quantitative relation between the effective conductivity versus grating period $\kappa_{\text{eff}}(\lambda)$ and cumulative conductivity versus phonon MFP $\kappa_{\Sigma}(\Lambda)$, which expresses the thermal conductivity governed by phonon modes having MFP $\leq \Lambda$, is far more complex than a simple one-to-one correspondence [8,11]. However, qualitative features remain reasonably preserved. We therefore see that materials exhibiting low/high $m$ values will be associated with MFP spectra $\kappa_{\Sigma}(\Lambda)$ having long/short tails (gradual/sharp convergence towards the bulk value $\kappa_0$). Based on this insight, it makes physical sense that the value we obtained in the main manuscript for the Si membrane ($m = 2$) is larger than for bulk GaAs ($m = 1.4$): thin film boundary scattering in the membrane affects long MFP modes the most severely, and essentially compresses the tail of the MFP spectrum.