Nonlocal Phonon Heat Transport Seen in 1-d Pulses

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Propagation of vibrational energy from an initial localized pulse gives useful insights into non-local phonon transport. We report simulations in one dimension. The 1d case has tricky anomalies, but provides the simplest pictures of the evolution from initially ballistic toward longer time diffusive propagation. Our results show interesting details, such as diverse results from different definitions of atomistic local energy, and failure to exhibit pure diffusion at long times. Boltzmann phonon gas theory, including external energy insertion, is applied to this inherently time-dependent and nonlocal problem. The solution, using relaxation time approximation for impurity scattering, does not closely agree with the simulated results.

I. INTRODUCTION

Figure 1 shows the local energy at atom sites on a one-dimensional chain of \( N = 200 \) atoms coupled by nearest-neighbor harmonic springs. The system has periodic boundary conditions. At time \( t = 0^- \), the chain is in its ground state. At \( t = 0 \), one or two central atoms are disturbed, starting a pulse. No further external disturbance is applied. The classical time evolution is computed from Newton’s laws. There are two main points of this paper. (1) To gain insight into the “crossover” from the ballistic propagation of Fig. 1 toward diffusive heat propagation when phonons start to evolve toward a local equilibrium \( T(r, t) \) because of scattering\(^{1,2} \). (2) To test the form and the accuracy of a non-local Boltzmann equation description\(^3 \).

The chain has atoms of mass \( M = 1 \) separated by distance \( a = 1 \) and connected by springs of constant \( K = 1 \). The harmonic normal modes have frequency \( \omega_Q = \omega_M \sin(Qa/2) \) where \( \omega_M = 2\sqrt{K/M} = 2 \). Time is measured in units \( \sqrt{M/K} = 2/\omega_M = 1 \). The modes propagate at velocities \( v_Q = v_M \cos(Qa/2) \text{sign}(Q) \) where \( v_M = \sqrt{K/M}a = 1 \). The unit of energy is \( E_0 = 1 = Ka^2 = Mv_M^2 = M\omega_M^2a^2/4 \). The leading edges of the pulse propagate at the speed of sound, \( \pm v_M = \pm 1 \), as is seen at \( t = 30 \) in Fig. 1.

The Hamiltonian of the chain is

\[
\mathcal{H} = \sum_\ell \left[ \frac{P_\ell^2}{2M} + \frac{1}{2}K(u_\ell - u_{\ell+1})^2 \right].
\]

(1)

Atoms have displacements \( u_\ell \) around average positions \( r_\ell = \ell a \). The general solution of Newton’s equations of motion is

\[
u_\ell(t) = \sqrt{\frac{1}{N}} \sum_Q A_Q \cos(Q\ell a - \omega_Q t + \phi_Q),
\]

(2)

There are \( 2N \) free parameters, amplitude \( A_Q \geq 0 \) and phase \( \phi_Q \) for each normal mode \( Q \). The wavevector has the form \( Q = (2\pi/a)(n/N) \), and the integer \( n \) lies in the range \(-N/2 < n \leq N/2 \). The pulses shown in Fig. 1 labeled D, V2, and V1, are generated by initial disturbances given in Table I. The “V1” (or “velocity”) pulse has only the central atom given a velocity \( v_0 \) at \( t = 0 \). The “V2” (or “dual velocity”) pulse has two central atoms given equal and opposite velocities. The “D” (or “displacement”) pulse has two central atoms given equal and opposite displacements at \( t = 0 \).

The first result to notice is the interesting diversity of pulse shapes for different initial disturbances, as was first noticed in ref. 4. The first issue to resolve is whether...
Local energy is well-defined.

II. DEFINING LOCAL ENERGY

Local energy $E(\ell, t)$ cannot be unambiguously defined. In classical physics it should obey $\sum_\ell E(\ell, t) = \mathcal{H}$. The version plotted in Fig. 1 is defined as

$$E_a(\ell, t) = \frac{M v_\ell(t)^2}{2} + \frac{K}{4} \left[ (u_{\ell-1}(t) - u_\ell(t))^2 + (u_\ell(t) - u_{\ell+1}(t))^2 \right].$$

(3)

Each atom is assigned its own kinetic energy, and half of the potential energy of the springs to its left and right. This is a commonly used definition, but it is not unique. Two other sensible choices for distributing potential energy to different sites are

$$E_b(\ell) = \frac{M v_\ell^2}{2} + \frac{K}{2} \left[ (2u_\ell^2 - u_{\ell-1} + u_{\ell+1}) \right],$$

(4)

$$E_c(\ell) = \frac{M v_\ell^2}{2} \quad \text{and} \quad E_c(\ell + 1/2) = \frac{V}{2} (u_\ell - u_{\ell+1})^2.$$  

(5)

The conventional version $E_a$ will be used in this paper, but it is interesting to see how it compares with versions $E_b$ and $E_c$, as shown in Fig. 2. The large oscillations seen in version (c) are surprising, and the smoothness seen in version (b) is even more surprising. This diversity illustrates our second result, a well-known fact: local energy at the atom level is not a clear concept. However, if local energy is averaged over a few nearby atoms, it becomes less diverse. The ambiguity also exists in quantum treatments. Marcolongo et al. and Ercole et al. have shown how this ambiguity does not affect the computation of bulk transport. In the next section we will see that local energy makes more sense in a continuum theory than an atomistic theory.

Figure 1 shows $E_a(\ell, t)$ for pulses, inserted at $t = 0$ into zero-temperature (i.e., stationary, $T = 0$) chains. Figure 2 shows the same pulse forms, at $t = 20$, inserted into chains with a pre-existing thermal distribution of velocities and displacements. In $T > 0$ cases, the initial pulse amplitudes ($\Delta u_{0,0}$ or $\Delta v_{0,0}$) are scaled from those in Table I to make the total extra energy $\sum_\ell \Delta E_a(\ell)$ of all pulses of the ensemble equal to 1.

![Figure 2](image)

**FIG. 2.** The harmonic V2 pulse at temperature $T = 0$ and time $t = 30$, using three ways (Eqs. 2) of assigning potential energy to a local site energy, $E(\ell)$. In (a) and (b), the pulse energy is $E_{\text{pulse}} = E_0 = \sum_\ell E(\ell)$. The area under the curves is $E_0$, chosen to be 1. In (c), however, the energy has two parts, $\sum_\ell \{KE(\ell) + PE(\ell + 1/2)\}$. The values in the graph have been arbitrarily doubled so that the total energy, also 1, is the area under the graph. These graphs demonstrate that the “continuum picture” of energy density (the dashed curves in Fig. 1) is more sensible than these local pictures.

Table I. Properties of pulses. The shift $\Delta u$ (in units of $a$) of initial displacement, or $\Delta v$ (in units of $v_{0x}$) of initial velocity, is scaled so that the new coordinates $u_\ell + \Delta u_0$, $v_\ell + \Delta v_0$, have total extra energy $E_{\text{pulse}} = 1$. The values in the table give $E_{\text{pulse}} = 0$ at $T = 0$, i.e. when $u_\ell(t) = 0$ for $t < 0$.

| name | $\Delta u_0$ | $\Delta u_1$ | $\Delta v_0$ | $\Delta v_1$ |
|------|-------------|-------------|-------------|-------------|
| V1   | 0           | 0           | $\sqrt{2}$  | 0           |
| V2   | 0           | 0           | -1          | 1           |
| D    | $-1/\sqrt{3}$ | 1/\sqrt{3} | 0           | 0           |

For the $T = 0$ case, values of $A_Q$ and $\phi_Q$ are given in Table II. They are derived from the $t = 0^+$ positions and velocities shown in Table I at $t = 0$.

The chains are harmonic, so the pulses propagate ballistically. The left or right parts have root mean square (rms) displacements $\bar{r}$ defined as

$$\bar{r}(t) = \left[ \frac{\sum_\ell (\ell a)^2 \Delta E_a(\ell, t)}{\sum_\ell \Delta E_a(\ell, t)} \right]^{1/2}.$$  

(7)

The rms displacements increase at speeds $v_{\text{rms}} = d\bar{r}/dt \approx v_M/\sqrt{n}$, with $n = 2, 4, 6$ for the V1, V2, and D pulses.
FIG. 3. Pulses of total energy $\sum_\ell \Delta E_\alpha(\ell) = E_0 = 1$ at $t = 20$, inserted at $t = 0$ to harmonic chains whose random positions and velocities at $t = 0$ are thermalized at temperature $T = 0.3$ ($E_{\text{tot}, \text{th}} = 0.3 N$). The local energies $\Delta E_\alpha(\ell)$ shown in the graphs are the differences between $E_\alpha(\ell, t = 20)$ with and without pulse insertion. The first graph shows D pulses inserted with two typical initial conditions. The other graphs show, in red, the results averaged over 1000-member ensembles with random thermal initial conditions. The black results repeat the zero-temperature pulses shown in Fig. 1. The finite $T$ pulse shapes, after ensemble averaging, are converging towards the zero-temperature pulse shapes.

After integrating over $Q$, the results for the three pulses are

$$\Delta E_{V1}(r, t) = \frac{E_0}{\pi v_M t} \left(1 - \left(\frac{r}{v_M t}\right)^2\right)^{1/2} \theta(v_M t - |r|),$$

$$\Delta E_{V2}(r, t) = \frac{2E_0}{\pi v_M t} \left(1 - \left(\frac{r}{v_M t}\right)^2\right)^{1/2} \theta(v_M t - |r|),$$

$$\Delta E_D(r, t) = \frac{8E_0}{3\pi v_M t} \left(1 - \left(\frac{r}{v_M t}\right)^2\right)^{3/2} \theta(v_M t - |r|).$$

The total energy $\int dr \Delta E(r, t)$ is $E_0$ in all three cases. These formulas are shown as dashed lines in Figs. 1 and 3. The continuum model agrees well with an average of nearby values of the local atomic energy $E(\ell)$ for all of the three pulse types $E_\alpha(\ell)$. The rms centers of energy of the propagating pulses are then

$$\bar{r}_{\text{continuum}}(t) \equiv \left[ \int dr r^2 \Delta E(r, t) \right]^{1/2} \int dr \Delta E(r, t).$$

This is where the result $d\bar{r}/dt \rightarrow v_M/\sqrt{n}$ (with $n = 2, 4, 6$) came from.

It had been our original guess that when the pulse propagates in a thermal background, the fine structure

| name | Amplitude $A_Q$ from Eq. 6 | phase $\phi_Q$ | modal energy $\Delta E^{\text{ext}}_Q$ |
|------|------------------|----------------|-------------------|
| V1   | $\frac{\Delta \rho_0}{\sqrt{N\omega_Q}}$ | $\frac{\pi}{2}$ | $\frac{E_0}{\pi}$ |
| V2   | $\frac{2\Delta \rho_0}{\sqrt{N\omega_Q}} \sin \left(\frac{Qa}{2}\right)$ | $\pi - \frac{Qa}{2}$ | $\frac{2E_0}{N} \sin^2 \left(\frac{Qa}{2}\right)$ |
| D    | $\frac{2\Delta \rho_0}{\sqrt{N\omega_Q}} \sin \left(\frac{Qa}{2}\right)$ | $\frac{\pi}{2} - \frac{Qa}{2}$ | $\frac{8E_0}{3N} \sin^4 \left(\frac{Qa}{2}\right)$ |

TABLE II. More properties of pulses: the distribution among normal modes $Q$ of the phonon amplitude $A_Q$, phase $\phi_Q$, and energy, for the pulses of Table I inserted at $T = 0$. respectively. These values are derived from the continuum description described next.

### III. A CONTINUUM DESCRIPTION

The energy content of each normal mode is

$$E(Q) = \frac{1}{2} M \omega_Q^2 A_Q^2.$$  \hspace{1cm} (8)

Using values of $A_Q$ from Table II, the mode energies are also shown in Table II. A continuum description uses spatially averaged atomic coordinates, and requires a new definition of local energy density $E(r, t)$. An appropriate definition for a pulse originating at $(r, t) = (0, 0)$ is

$$\Delta E(r, t) = \sum_Q \Delta E^{\text{ext}}_Q \delta(r - v_Q t).$$  \hspace{1cm} (9)

The total energy $\int dr \Delta E(r, t)$ is $E_0$ in all three cases. These formulas are shown as dashed lines in Figs. 1 and 3. The continuum model agrees well with an average of nearby values of the local atomic energy $E(\ell)$ for all of the three pulse types $E_\alpha(\ell)$. The rms centers of energy of the propagating pulses are then

$$\bar{r}_{\text{continuum}}(t) \equiv \left[ \int dr r^2 \Delta E(r, t) \right]^{1/2} \int dr \Delta E(r, t).$$  \hspace{1cm} (13)
in $E(\ell)$ would disappear and the result would resemble the continuum version $E(r,t)$. The computation in Fig. 3 shows that this guess was wrong. The fine structure remains. A proof that this should happen is given in the Supplemental Material.\(^{[3]}\)

**IV. NON-LOCAL BOLTZMANN EQUATION, COLLISIONLESS**

The formulas given in Eqs. \(^{[10]}\) \(^{[11]}\) \(^{[12]}\) came from a sensible hypothesis, Eq. \(^{[9]}\) which will now be derived from Boltzmann theory. Pulse behavior is fundamentally non-local, and not closely associated with deviation of temperature $T(r,t)$ from background $T_0$. The usual local Boltzmann equation is very successful\(^{[2]}\) in describing the bulk law $j = -\kappa \nabla T$, where the temperature gradient $\nabla T$ is constant in space and time, or varying slowly on the scale of phonon mean free paths $\ell_Q$ and lifetimes $\tau_Q$. Pulse propagation requires an extension of the usual local version. For at least 60 years\(^{[10]}\)\(^{[12]}\) there have been developments of Boltzmann theory aimed at studying systems driven at small distance scales, i.e. distances comparable to quasiparticle mean free paths. There are many recent studies, for example, refs. \(^{[3]}\) \(^{[4]}\) \(^{[13]}\) \(^{[20]}\). There are also models outside of Boltzmann theory that apply when geometries are too complex.\(^{[21]}\)

The Peierls Boltzmann equation\(^{[22]}\) (PBE) uses quantum wave/particle duality to describe the system as a gas of phonon particles in a continuous space with coordinate $r$, rather than discrete sites $r_i = l\ell$ on a lattice. Phonons can be treated either as classical or quantum particles. A correct treatment of Boltzmann theory with the full scattering operator $(\partial N_Q/\partial t)_{\text{coll}}$ gives the same low frequency (and spatially homogeneous) transport properties as a self-consistent treatment by Green-Kubo theory to second order in interactions.\(^{[13]}\)\(^{[22]}\) The fundamental object is $N_Q(r,t)$, the occupancy per unit volume $V$ (where $V = \text{length } L$ in 1d) of phonon mode $Q$ at $(r,t)$. The spatial sum $\int dr N_Q(r,t) = N_Q(t)$ is the mode occupancy. The PBE is

$$
\frac{\partial N_Q}{\partial t} = -v_Q \frac{\partial N_Q}{\partial r} + \left( \frac{\partial N_Q}{\partial t} \right)_{\text{coll}} + \left( \frac{\partial N_Q}{\partial t} \right)_{\text{ext}}.
$$

(14)

Local energy is $E(r,t) = (1/V) \sum_i \hbar \omega_Q N_Q(r,t)$. Heat current density is $j(r,t) = (1/V) \sum_i \hbar \omega_Q v_i N_Q(r,t)$. Local energy is conserved. Summing Eq. (14) over $Q$ (after multiplying by $\hbar \omega_Q$) gives

$$
\frac{\partial E(r,t)}{\partial t} = -\nabla j(r,t) + \left( \frac{\partial E(r,t)}{\partial t} \right)_{\text{ext}}.
$$

(15)

This holds because collisions conserve energy locally, $\sum_i \hbar \omega_Q (\partial N_Q/\partial t)_{\text{coll}} = 0$. We will use the quantum version, with an equilibrium Bose-Einstein distribution $N_Q \rightarrow n_Q(T_0) = [\exp(\hbar \omega_Q/k_B T_0) - 1]^{-1}$, and take the classical limit $n_Q(T) = k_B T / \hbar \omega_Q$ when comparing with simulations.

The scattering (or collision) term $(\partial N_Q/\partial t)_{\text{coll}}$ tries to drive the local distribution to a local thermal distribution. Writing the distribution as $n_Q(T(r,t)) + \Phi_Q(r,t)$, where $\Phi_Q$ is the deviation from local equilibrium, and linearizing, Eq. (14) becomes

$$
\frac{\partial n_Q}{\partial t} + \frac{\partial \Phi_Q}{\partial t} = -v_Q \left( \frac{\partial n_Q}{\partial T} \frac{\partial T}{\partial r} + \frac{\partial \Phi_Q}{\partial r} \right) - \sum_{Q'} C_{QQ'} \Phi_{Q'} + \left( \frac{\partial N_Q}{\partial t} \right)_{\text{ext}}.
$$

(16)

where $-C_{QQ'}$ is the linearized scattering operator. The atoms are driven by external manipulation that changes the Newtonian state $\{u_i, v_i\}$ to $\{u_i + \Delta u_i, v_i + \Delta v_i\}$ at $t = 0$, for $\ell = 0$ and 1. It changes the phonon amplitudes and phases to give the starting pulse shape. A continuum version of the change must be created by the term $(\partial N_Q/\partial t)_{\text{ext}}$ in the Boltzmann equation. External driving $(\partial N_Q/\partial t)_{\text{ext}}$ has only recently appeared in phonon Boltzmann theory.\(^{[21]}\)\(^{[25]}\)\(^{[29]}\) Whether its form and significance is still open to discussion. Boltzmann theory does not deal directly with amplitudes $A_Q$. These are indirectly included via the mode energy $MA^2_Q \omega_Q^2 / 2 \rightarrow \hbar \omega_Q N_Q(1/2)$. Coherent phase relations $\phi_Q$ between different quasiparticles $Q$ cannot be handled. A choice for the external term $(\partial N_Q/\partial t)_{\text{ext}}$ driving the distribution function $N_Q$ away from equilibrium is

$$
\left( \frac{\partial N_Q}{\partial t} \right)_{\text{ext}} = \frac{\Delta E_{Q}^{\text{ext}}}{\hbar \omega_Q} \delta(r) \delta(t).
$$

(17)

A very similar version of Boltzmann theory applied to time-domain thermoreflectance was given in ref. \(^{[2]}\). The energy inserted by the pulse into mode $Q$ is $\hbar \omega_Q \Delta N_Q^{\text{ext}}$, where $\Delta N_Q^{\text{ext}} = \int dr \int dt (\partial N_Q(r,t)/\partial t)_{\text{ext}}$. The total pulse energy given to the system is clearly correct:

$$
E_{\text{pulse}}^{\text{Boltzmann}} = \sum_Q \Delta E_{Q}^{\text{ext}}.
$$

(18)

Because of linearity and periodic boundary conditions, it is convenient to Fourier transform to $N_Q(k,\omega)$,

$$
N_Q(k,\omega) = \frac{1}{L} \int_{-L/2}^{L/2} dr \int_{-\infty}^{\infty} dt N_Q(r,t)e^{-i(kr-\omega t)}.
$$

(19)

Equation (16) becomes

$$
\sum_{Q'} C_{QQ'} \Phi_{Q'} + i(kv_Q - \omega) \Phi_Q = \frac{-i(kv_Q - \omega) \frac{\partial n_Q}{\partial T} \Delta T(k,\omega) + \Delta E_{Q}^{\text{ext}}}{\hbar \omega_Q}.
$$

(20)

In the harmonic case, there are no collisions and $C_{QQ'} = 0$. The solution in Fourier space is

$$
\frac{\partial n_Q}{\partial T} \Delta T(k,\omega) + \Phi_Q(k,\omega) = \frac{\Delta E_{Q}^{\text{ext}}}{i(kv_Q - \omega - \eta)}.
$$

(21)
The left hand side is $\Delta N_Q(k, \omega) = N_Q(k, \omega) - n_Q(T_0)$. Transforming back to $(r, t)$-space,

$$
\Delta N_Q(r, t) = \frac{L}{2\pi} \int dk \int d\omega \frac{\Delta E^{\text{ext}}_Q/\hbar \omega}{2\pi - i(\omega + i\eta - k v_Q)} e^{i(kr - \omega t)}
$$

$$
= \frac{\Delta E^{\text{ext}}_Q}{\hbar \omega} \delta(r - v_Q t)
$$

(22)

The local energy density is

$$
E_{\text{pulse}}(r, t) = \sum_Q \hbar \omega \Delta N_Q(r, t) = \sum_Q \Delta E^{\text{ext}}_Q \delta(r - v_Q t).
$$

(23)

This agrees exactly with Eq. [3]. These results provide confidence in the insertion term added to the Peierls Boltzmann equation. We also learn that, in the continuum description, harmonic pulse shapes (Eqs. [10–12]) are independent of $T$, because the temperature $T_0$ in the Boltzmann treatment did not have to be specified. The less obvious result, that harmonic pulse shapes in the atomistic version are also independent of $T$, is explained in the Supplemental Material.$^5$

V. MASS DISORDER

Disorder adds an interesting complication to heat conduction in 1-d harmonic crystals$^{10,11}$, namely Anderson localization.$^{33}$ In disordered metals of dimension 2 or less, ignoring electron-electron interactions, all single-particle electron eigenstates are localized.$^{33}$ At $T = 0$, an electron inserted into a localized state cannot propagate. Localization of phonons is similar$^{32,33}$ except that small $Q$ acoustic phonons have to travel very long distances before localization appears.$^{37}$ Reference $38$ gives the example of a wave packet propagating on a weakly mass-disordered chain. Ballistic propagation is seen at short distances and times, diffusive propagation at intermediate ones, and Anderson localization at long distances and times. When $T > 0$, interactions with phonons allow a localized electron to hop to neighboring localized states, which causes slow diffusion. Anharmonic interactions have a similar effect$^{32}$ on localized phonons in insulators. If disorder is not too great, phonon quasiparticles are a realistic model at intermediate times and distances. Ballistic phonons eventually scatter from disorder and evolve toward diffusive at moderate to long distances and times, before localizing. A perturbative treatment of scattering can likely describe the parts before localization sets in.

We now add mass defects to allow deviation from ballistic propagation, by randomly choosing 10% of the atoms, and increasing their masses from $M = 1$ to $M^* = 1.5$. Results at various times for a D pulse are shown in Fig. [4]. The lattice is still harmonic, but the Hamiltonian is no longer diagonal in the plane-wave basis, Eq. [2]. Before ensemble averaging, the pulse shape varies depending on the locations of the mass defects relative to the point of pulse insertion. The D-pulse shapes of Fig. [4] have been averaged over 100 different random placements of the altered masses.

The pulse shape at $t = 10\sqrt{M/\bar{k}}$ is not much altered from pure ballistic behavior. The pulse has propagated only a distance of $\pm 10$ atoms, and encountered typically only two impurities. As time proceeds, there is increasing deviation from the ballistic pulse shape predicted in Eq. [12] and shown in the red curves. By $t = 40$, the fraction of the energy at distances $< 10a$ has failed to diminish the way ballistic propagation does. The energy in the intermediate $10 - 30$ atoms has diminished more than ballistic propagation does. The crossover toward diffusion is underway. The disorder is weak, so the Anderson localization lengths $\xi_Q$ are mostly longer than the propagation distance ($\leq 40a$) studied here.

VI. PURE DIFFUSION

The opposite extreme from ballistic propagation is pure diffusion. The equation for the energy $\Delta E$ propagating diffusively from a pulse $P$ is

$$
\left( \frac{\partial}{\partial t} - D \frac{d^2}{dr^2} \right) \Delta E(r, t) = P(r, t) = E_0 \delta(r) \delta(t)
$$

(24)

This follows from energy conservation, Eq. [15] provided there is a local relation $j = -\kappa d\bar{T}/dr$ between current $j(r, t)$ and temperature $\bar{T}(r, t)$. It also uses as the definition of temperature $\Delta E(r, t) = CV \Delta T(r, t)$. Then the diffusion constant is $D = \kappa/C$ where $\kappa$ and $C$ are bulk thermal conductivity and specific heat. The solution of Eq. [24] is

$$
\Delta E_{\text{diff}}(r, t) = \frac{E_0}{\sqrt{4\pi D t}} e^{-r^2/4Dt} \theta(t),
$$

(25)

where $\theta(t)$ is the Heaviside function.

Pure diffusion is inconsistent with a quasiparticle picture of pulse evolution. One argument is that it violates the rule that lattice vibrational energy cannot propagate faster than the speed of sound $v_M$. It is not necessarily a large violation. An estimate of the size uses $\kappa \approx Cv \ell = C g^2 \tau$. The diffusive exponent $r^2/4Dt$ is then approximately $(\ell/4\tau)(r/v_M t)^2$ (where $\ell$ and $\tau$ are rough measures of mean free path and lifetime of phonons). Therefore, when $t = 4\tau$, there is some diffusive energy at $r > v_M t$, which decays rapidly as $r/v_M t$ or $t/4\tau$ increases.

Another argument for the inapplicability of pure diffusion to quasiparticles is that mean free paths of small $Q$ acoustic phonons typically diverge as a power, $\ell_Q \propto 1/Q^p$, causing $D$ to diverge. This is a correct result, not an error of perturbation theory. A formula for $D = \kappa/C$ is found from the standard RTA solution of Boltzmann theory for $\kappa$ in the bulk limit,

$$
D = \frac{\sum Q C_Q v_Q^2 \tau_Q}{\sum Q C_Q} \to \frac{1}{N} \sum Q v_Q^2 \tau_Q.
$$

(26)
The second form is the classical limit where \( C_Q = \hbar \omega_Q \delta_{0Q} / dT \to k_B \). When \( 1/\tau_Q \) arises from mass disorder, the small \( Q \) scattering rate \( 1/\tau_Q \) in \( d = 1 \) goes as \( Q^2 \)\cite{30,30,30,30}, shown explicitly in appendix A. The divergence is not limited to one dimension. Small \( Q \) scattering from mass defects is closely analogous to Rayleigh scattering of light from density fluctuations. In 3d, both light and phonon scattering have \( Q^4 \) dependences at small \( Q \). The \( Q \)-sum needed to compute \( D \) or \( \kappa \) diverges as \( 1/Q^2 \) in both \( d = 1 \) and \( d = 3 \), unless another scattering process adds a term to \( 1/\tau_Q \) that blocks the divergence. When \( T \) is small enough that mean free paths \( \ell_Q = v_Q \tau_Q \) reach sample dimensions \( L \), the zero denominator from the diverging defect term becomes non-zero due to boundary scattering \( L = v_Q \tau_{\text{boundary}} \). Glassbrenner and Slack analyze experiments which illustrate this\cite{44,44,44,44}. It is seen in clean but isotopically disordered crystals\cite{30,30,30,30}. Eqn. 26 can be replaced by

\[
D = \sum_Q \left[ \left( 1/\tau_Q \right)_{\text{disorder}} + |v_Q|/L \right]^{-1}.
\]

The sum now converges, diverging for large \( L \) as \( D \propto \sqrt{L} \) in \( d = 1 \). The \( \sqrt{L} \) scaling of \( \kappa \) in \( d = 1 \) was noticed earlier\cite{30,30,30,30}. The resulting \( D \) is plotted versus \( L/a \) in Fig. 5. Our Newtonian simulation on a loop of 200 atoms has no boundary scattering because of periodic boundary conditions. If we had used instead rough boundaries, then \( L \) in Eq. 27 would be \( 100a \), resulting in \( D = 35\omega_M a^2/2 \) (see Appendix A).

In spite of the inapplicability of Eq. 25, it is interesting to compare it to the numerical results. Figure 6 compares the simulation result of the disordered \( D \)-pulse at \( t = 40 \) (the last graph of Fig. 4) with the formulas for pure ballistic and pure diffusive propagation. For the majority of atoms (all but atoms 10-14) the computed pulse energy is closer to the green curve illustrating pure diffusion (with \( D = 1.8 \)) than to the red curve of pure ballistic behavior. However, the agreement with pure diffusion is poor, and more important, the choice \( D = 1.8 \) was chosen to give a curve for \( \Delta E(r, t = 40) \) reasonably close to the simulation, but it is totally unrealistic, corresponding to a nanoscale sample with boundaries at \( L \approx \pm 3a \), as seen from Fig. 5.

VII. BOLTZMANN THEORY WITH MASS DISORDER

How does non-local Boltzmann theory treat pulse shape evolution as altered by disorder? Here we answer this question within the relaxation time approximation (RTA), and find that the results have the correct trend but do not agree closely with simulations. The collision operator \( C_{QQ'} \) in Eq. 20 is replaced in RTA by \( 1/\tau_Q \delta_{QQ'} \),
The requirement of energy conservation says that
\[
\sum_Q \hbar \omega_Q (d\Phi_Q/dt)_{\text{coll}} = 0.
\]
The correct scattering operator satisfies this automatically, but the RTA version,
\[
(dE/dt)_{\text{coll,RTA}} = -\sum_Q \hbar \omega_Q \Phi_Q / \tau_Q = 0,
\]
is not automatically satisfied. Forcing \( \Phi_Q \) to satisfy this equation as well as the linearized PBE is one way to obtain the extra equation needed to determine \( \Delta T(k,\omega) \) and \( \Delta T(r,t) \). This definition of temperature which we call “version (1)”, is not a normal one. A possible alternative is to define temperature in terms of the local energy, \( E(r,t) = \sum_Q \hbar \omega_Q n_Q(T(r,t)) \) or \( E(k,\omega) = \sum_Q \hbar \omega_Q (dn_Q/dT) \Delta T(k,\omega) \). This, called “version (2)”, is the definition of \( \Delta T(k,\omega) \) used in the full Boltzmann equation with the correct scattering operator. It requires

the deviation \( \Phi_Q(r,t) \) to have no net energy. This definition of temperature is quite normal - it is sensible in a quasiparticle theory, although perhaps not demanded by nonequilibrium thermodynamics.

The two possible versions of the extra equation, needed in RTA, are then

\[
(1) \sum_Q \hbar \omega_Q \Phi_Q, \text{RTA} / \tau_Q = 0; \text{ or } (2) \sum_Q \hbar \omega_Q \Phi_Q, \text{RTA} = 0.
\]

We find that version (1) gives a more realistic answer, in agreement with previous numerical work.

The pulse energy predicted by version (1) is shown in Fig. 6. It deviates less from ballistic than does the simulation, but in the correct direction. Energy profiles predicted by both versions of the RTA Boltzmann theory are in Fig. 6 for the D pulse at \( t = 40 \). Details of the computational procedure are given in appendix B and in the Supplemental Material. Version (1) of RTA theory correctly keeps the total pulse energy equal to \( E_0 \) as \( t \) increases, while version (2) does not. Version (2) has another weakness, namely the predicted shape of the evolving pulse in Fig. 7 is very different from the simulated results in Fig. 6, unlike version (1) where the Boltzmann-RTA pulse shape is sensible. However, version (1) has a temperature profile \( \Delta T(r,t) \) in Fig. 7 that differs from \( \Delta E(r,t) \) without physical justification, unlike version (2) which correctly equates \( k_B T(r,t) \) and \( \Delta E(r,t) \).
Heat conduction in weakly anharmonic linear lattices has been studied\cite{22} and reviewed\cite{22}. Recent work\cite{23} seems to confirm (at least qualitatively) that perturbation theory works in the regime studied here. If so, the perturbative picture says that mean free paths will diminish as $T$ increases.\cite{24}\cite{25} A crossover to diffusive behavior, which FPU indicates might not happen at $T = 0$, may well happen at $T > 0$, at rates that increase as $T$ increases.

We have looked at $T = 0$ anharmonic D-pulse propagation, using both third-order ($V_3$, also known as “FPU–α”) and fourth-order ($V_4$, also known as “FPU–β”) anharmonicity. The coefficients $\alpha$ and $\beta$ in Eqs. \cite{31} and \cite{32} were set to 1. Local energy $E(\ell,t)$ is defined as in Eq. \cite{3} the total (harmonic and anharmonic) potential energy of a spring is assigned half to each neighboring atom. The results are in Fig. \ref{fig:Fig. 8}. The initial pulse ($\varphi_{0,1} = \pm 0.3$) is the same as in previous computations, except the total energy is no longer $0.27E_0$ as in previous computations, because there is additional anharmonic energy, $-27\%$ in $V_3$ and $+12\%$ in $V_4$. The initial pulse has the same harmonic amplitudes $A_Q$ and phases $\phi_Q$ as previously, but anharmonic terms alter these fairly quickly. As the pulse spreads and $\Delta E(\ell,t)$ spreads out, anharmonic forces diminish. Amplitudes and phases evolve less, becoming more stable. As $t$ increases, local energy propagation from $(x,t)$ reverts more closely to ballistic. This is especially noticeable in the $V_3$ case, where amplitudes $10 - 16$ seem little affected by anharmonic effects. The $V_5$ case might be especially difficult to analyze perturbatively, because in lowest order and 1d, anharmonic decay is essentially kinematically forbidden, requiring higher order effects to change $A_Q$ and $\phi_Q$. It would be interesting to study the anharmonic pulse at finite $T$ where anharmonic effects do not disappear as the pulse propagates.\cite{26}

\section{Conclusions}

Time evolution of pulse energy gives useful pictures and insights into non-local phonon transport. The main conclusions are: (1) Different forms of pulse insertions give interestingly diverse pulse energy shapes. (2) Atomistic local energy $E(\ell,t)$ is not uniquely defined and has surprisingly different details when different sensible definitions are used. (3) A continuum picture works well and enables simple formulas for ballistic propagation. (4) A non-local version of Boltzmann theory for the collisionless phonon gas is very successful, and the phonon insertion term $(\partial N_Q/\partial t)_{\text{ext}}$ has an unambiguous form. (5) Mildly disordered harmonic systems have interesting pulse evolution, but are not well explained by non-local Boltzmann theory in relaxation time approximation (RTA). Ambiguity about temperature definition in RTA is a difficulty;
previous ideas are confirmed. (6) Pure diffusion does not work at the local level, when phonon quasiparticles are good approximations, even after long pulse evolution times. (7) The pulse evolution of 1d anharmonic chains at $T > 0$ deserves further study.

X. ACKNOWLEDGEMENTS

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Appendix A: Phonon relaxation from mass disorder

The Fermi golden rule for phonon decay by defects is $1/\tau_Q = (2\pi/\hbar)\sum_{Q'}|V_{QQ'}|^2\delta(\omega_Q - \omega_{Q'})$. When the interaction $V_{QQ'}$ arises from mass disorder, the decay rate is

$$\left(\frac{1}{\tau_Q}\right)_{\text{imp}} = \frac{\pi}{2\hbar} N_i \left(\frac{\Delta M}{M^*}\right)^2 (\hbar\omega_Q)^2 \mathcal{D}(\omega_Q),$$

(A1)

where $\Delta M = M^* - M$ is the mass difference between impurities ($M^*$) and pure ($M$) masses, and $N_i/N$ is the fraction of impurities. The density of vibrational states of the ordered harmonic chain is

$$\mathcal{D}(\omega) = \frac{1}{\hbar N} \sum_Q \delta(\omega - \omega_Q) = \frac{2}{\pi\hbar} \frac{1}{(\omega_Q^2 - \omega^2)^{1/2}}.$$  

(A2)

Then the decay rate is

$$\frac{1}{\tau_Q} = \epsilon \frac{\omega_Q^2}{(\omega^2 - \omega_Q^2)^{1/2}}, \quad \text{where} \quad \epsilon = \frac{N_i}{N} \left(\frac{\Delta M}{M^*}\right)^2,$$

(A3)

and $\epsilon = 1/90$ in our simulations.

The formula[27] for boundary-limited diffusivity can be written as

$$D(L) = \frac{\omega_M a^2}{2} s \frac{\pi}{\epsilon} \int_0^{\pi/2} dx \frac{\cos^3 x}{\cos^2 x + s \sin^2 x},$$

(A4)

where $s = 2L\epsilon/a$ and $\omega_M a^2/2 = 1$ is the unit of diffusivity. At large $s$, the diffusivity scales as $\sqrt{s/\epsilon} \propto \sqrt{L/\epsilon}$.

Appendix B: Solution of Boltzmann equation in RTA

Using Eq. 29 and version (1) of Eq. 30 the equation for $\Delta T$ is

$$k_B \Delta T_{(1)}(k,\omega) = \frac{A(k,\omega)}{B(k,\omega)},$$

(B1)

where

$$A(k,\omega) = \frac{1}{N} \sum_Q \frac{\Delta E_{Q}^{\text{ext}}/\tau_Q}{1/\tau_Q - i(\omega - kv_Q)},$$

(B2)

$$B(k,\omega) = \frac{1}{N} \sum_Q \frac{i(\omega - kv_Q) [\hbar\omega_Q / k_B \partial_q]}{1/\tau_Q - i(\omega - kv_Q)}.$$  

(B3)

Once $\Delta T_{(1)}(k,\omega)$ is computed, the corresponding local energy $\Delta E_{(1)}(k,\omega)$ can be found and Fourier transformed to get $\Delta E_{(1)}(r,t)$,

$$\Delta E_{(1)}(k,\omega) \equiv \sum_Q \hbar\omega_Q [N_Q - n_Q(T_0)]$$

$$= C(k,\omega) k_B \Delta T_{(1)}(k,\omega) + D(k,\omega).$$  

(B4)

where

$$C(k,\omega) = \frac{1}{N} \sum_Q \frac{1/\tau_Q [\hbar\omega_Q / k_B \partial_q]}{1/\tau_Q - i(\omega - kv_Q)},$$

(B5)
\[ D(k, \omega) = \frac{1}{N} \sum_Q \Delta E^\text{ext}_Q \frac{\Delta E^\text{ext}_Q}{1/\tau_Q - i(\omega - kv_Q)} . \] (B6)

The factor in square brackets ([ ]) in Eqs. B3 and B5 (and later in Eq. B9) becomes 1 in the classical limit, which is needed for comparison with the numerical pulse spreading.

To implement version (2), use the simpler condition

\[ \Delta E_{(2)}(k, \omega) = k_B \Delta T_{(2)}(k, \omega) . \] (B7)

Using Eq. 29 and version (2) of Eq. 30 the equation for \( \Delta E \) is

\[ \Delta E_{(2)}(k, \omega) = k_B \Delta T_{(2)}(k, \omega) = \frac{D(k, \omega)}{F(k, \omega)} , \] (B8)

where, except for omission of factors of 1/\( \tau_Q \) in the numerator, \( D(k, \omega) \) is the same as \( A(k, \omega) \) and \( F(k, \omega) \) is the same as \( B(k, \omega) \). To be explicit,

\[ F(k, \omega) = \frac{1}{N} \sum_Q -i(\omega - kv_Q) \left[ \frac{h\omega \partial_{n^Q}}{k_B} \frac{\partial T}{\partial r} \right] \frac{1/\tau_Q - i(\omega - kv_Q)}{1/\tau_Q - i(\omega - kv_Q)} . \] (B9)

Details of the numerical calculations, especially the difficult Fourier transforms needed to get pulse shapes in (\( x, t \))-space, are in the Supplemental Materials.

**Appendix C: Laplace transform method of Vermeersch et al.**

Vermeersch et al. (Ref. 2) used mixed Fourier (for space variables \( r \leftrightarrow k \)) and Laplace (for time variables \( t \leftrightarrow \omega \)) transforms to solve for the case of a V1 pulse in one dimension. They used version (1) of Eq. 30, but did not transform from \((k, \omega)\) to \((r, t)\). However, they found interesting identities in Laplace space, which we will here pursue for arbitrary \( E^\text{ext}_Q \), not just the V1 choice \( E^\text{ext}_Q = E_0/N \). The basis for their identities are the Fourier equations

\[ \frac{1}{N} \sum_r F(r, t) = [F(k, t)]_{k=0} , \] (C1)

\[ \frac{1}{N} \sum_r r^2 F(r, t) = - \left[ \frac{d^2}{dk^2} F(k, t) \right]_{k=0} . \] (C2)

Together, these give a result for a mean square displacement

\[ \langle r^2 \rangle_F = \frac{\sum_r r^2 F(r, t)}{\sum_r F(r, t)} , \] (C3)

where \( F \) is an arbitrary distribution.

When the time variable is transformed to Laplace space rather than Fourier space, \( \Phi_Q(r, \omega) \) becomes \( \int_0^\infty dt \exp(-\omega t) \Phi_Q(r, t) \). The symbol \( \omega \) previously used for the Fourier variable is now used for the Laplace variable. The solution of the Boltzmann equation is then exactly the same as previously, Eqs. B3 and B4, except that the Fourier variable \(-i\omega\) becomes the Laplace variable \( \omega \). Using the fact that sums over \( Q \) contains pairs \( Q \) and \( -Q \) with \( v_Q = -v_Q \), which causes imaginary parts to vanish, and using the notations \( C_Q = h\omega q\partial n_Q/\partial T \) and \( \Lambda_Q = v_Q \tau_Q \), the functions that determine \( \Delta T(k, \omega) \) and \( \Delta E(k, \omega) \) are

\[ A(k, \omega) = \frac{1}{N} \sum_Q \frac{\Delta E^\text{ext}_Q (1 + i\omega \tau_Q)}{(1 + \omega \tau_Q)^2 + (k\Lambda_Q)^2} , \] (C4)

\[ B(k, \omega) = \frac{1}{N} \sum_Q \frac{C_Q}{\tau_Q} \left[ 1 - \frac{1 + i\omega \tau_Q}{(1 + \omega \tau_Q)^2 + (k\Lambda_Q)^2} \right] , \] (C5)

\[ C(k, \omega) = \frac{1}{N} \sum_Q \frac{C_Q (1 + i\omega \tau_Q)}{(1 + \omega \tau_Q)^2 + (k\Lambda_Q)^2} , \] (C6)

\[ D(k, \omega) = \frac{1}{N} \sum_Q \frac{\Delta E^\text{ext}_Q \tau_Q (1 + i\omega \tau_Q)}{(1 + \omega \tau_Q)^2 + (k\Lambda_Q)^2} . \] (C7)

Then recalling that \( \Delta T(k, \omega) = A(k, \omega)/B(k, \omega) \), Eqs. C4 and C5 give Eq. 4 of Vermeersch et al. in the V1 case. They use \( C_Q \rightarrow k_B \), the classical limit, and set \( E^\text{ext}_Q = E_0 \rightarrow 1 \) (which they call a unit pulse). The general answer for the integrated temperature rise of a pulse is

\[ d_T(\omega) = \frac{1}{N} \sum_r \Delta T(r, \omega) = \frac{1}{\omega} \sum_Q \frac{\Delta E^\text{ext}_Q}{C_Q 1 + \omega \tau_Q} . \] (C8)

For the V1 pulse in the classical limit, this gives the total pulse temperature rise as \( \sum_r k_B \Delta T(r, \omega) = E_0/\omega \). Then the inverse Laplace transform gives the result of Ref. 2. \( \sum_r k_B \Delta T(r, t) = E_0 \). However, for other pulse forms of \( E^\text{ext}_Q \), there is no analytic inverse Laplace transform of Eq. C8. Also, Vermeersch et al. incorrectly identify \( \sum_r k_B \Delta T(r, t) \) with the pulse energy \( \sum_r \Delta E(r, t) \). This identification is version (2) of Eq. B9, but is inconsistent with version (1) which they (sensibly) adopt as the preferred RTA approximation. Fortunately, the erroneous identification of \( \Delta E(r, t) \) with \( k_B \Delta T(r, t) \) goes away in the V1 case when summed over all \( r \). Using the correct RTA version

\[ \Delta E(k, \omega) = k_B \frac{A(k, \omega)}{B(k, \omega)} C(k, \omega) + D(k, \omega) , \] (C9)

gives the result

\[ \Delta E(k = 0, \omega) = \frac{1}{\omega N} \sum_Q \Delta E^\text{ext}_Q = \frac{E_0}{\omega} \] (C10)
for all $\Delta E^\text{ext}_{Q}$, not just the V1 case. Doing the inverse Laplace transform then shows that the RTA in version (1) correctly conserves the pulse energy in time,

$$\sum_{r} \Delta E(r,t)_{\text{RTA},(1)} = E_{0}, \quad (C11)$$

not just for the V1 pulse, but for arbitrary choice of $\Delta E^\text{ext}_{Q}$. This is not surprising; version (1) of Eq. (30) enforces overall energy conservation in RTA. Version (2) does not and does not obey Eq. (C11). The Vermeersch et al. identification of $\Delta E(k,\omega)$ with $k_{B}\Delta T(k,\omega)$ is correct in the $k = 0$ limit, but only for the V1 pulse, not for others.

Now examine the mean square displacements, using Eq. (C2). For the case $\Delta T(k,\omega) = A(k,\omega)/B(k,\omega)$, the answer in Laplace space is

$$n_{T}(\omega) \equiv \frac{1}{N} \sum_{r} r^{2} \Delta T(r,\omega) = 2 \sum_{Q} \frac{\Delta E^\text{ext}_{Q} \Delta^{2}_{Q}}{(1 + \omega \tau_{Q})^{2}} \sum_{Q} \frac{C_{Q} \Lambda_{Q}^{2}}{(1 + \omega \tau_{Q})} + 2 \frac{\sum_{Q} \Delta E^\text{ext}_{Q} \Delta^{2}_{Q}}{(1 + \omega \tau_{Q})^{2}} \sum_{Q} \frac{C_{Q} \Lambda_{Q}^{2}}{(1 + \omega \tau_{Q})}
$$

(C12)

For the V1 pulse this simplifies to

$$\frac{1}{N} \sum_{r} r^{2} \Delta T(r,\omega) = \frac{2}{\omega^{2}} \sum_{Q} \frac{C_{Q} \Lambda_{Q}^{2}}{(1 + \omega \tau_{Q})^{2}}, \quad (C13)$$

which is Eq. 6 of Vermeersch et al.

The point of these calculations is that in principle we could do inverse Laplace transforms to get $n_{T}(t)$ from $n_{T}(\omega)$ and $d_{T}(t)$ from $d_{T}(\omega)$, and then take their ratio $n_{T}(t)/d_{T}(t)$ to get the mean square displacement $(r^{2}(t))_{T}$ of the pulse temperature profile $\Delta T(r,t)$. Analytic inversions are not available. Vermeersch et al. instead look at the limit of high $\omega \tau_{Q}$ which should give $\langle r^{2}(t)\rangle_{T}$ at small times $t$, and also at the limit of low $\omega \tau_{Q}$ which should give $\langle r^{2}(t)\rangle_{T}$ at large times $t$.

At large $\omega \tau_{Q}$, the $\omega$-dependences of $n_{T}(\omega)$ and $d_{T}(\omega)$ are

$$\frac{1}{N} \sum_{r} r^{2} \Delta T(r,\omega) \to \frac{2}{\omega^{3}} \sum_{Q} \frac{\Delta E^\text{ext}_{Q} \Delta^{2}_{Q}}{(1 + \omega \tau_{Q})^{2}} \sum_{Q} \frac{C_{Q}}{\tau_{Q}} + O\left(\frac{1}{\omega^{4}}\right), \quad (C14)$$

The inverse Laplace transform of $2/\omega^{3}$ is $t^{2}$, and of $1/\omega$ is 1. Then we get ballistic behavior in the short time limit, $\langle r^{2}\rangle_{T} \to \langle v^{2}\rangle_{T} t^{2}$

(C16)

where the mean square velocity of the temperature pulse is

$$\langle v^{2}\rangle_{T} \to \sum_{Q} \frac{\Delta E^\text{ext}_{Q} \Delta^{2}_{Q}}{\tau_{Q}} \sum_{Q} \frac{C_{Q}}{\tau_{Q}} \sum_{Q} \frac{\Delta E^\text{ext}_{Q}}{\tau_{Q}} \langle \sum_{Q} \frac{\Delta E^\text{ext}_{Q} \Delta^{2}_{Q}}{\tau_{Q}} \rangle + O\left(\frac{1}{\omega^{4}}\right). \quad (C17)$$

This agrees with Eq. 8 of Vermeersch et al. in the V1 case. It also resembles our result for the collisionless energy pulse propagation, namely Eqs. (9) or (23) plus Eq. (13). However, in contrast with the collisionless limit, there is an extra $T$-dependent factor $1/\tau_{Q}$ in the weights of $\langle v^{2}\rangle_{T}$ in Eq. (C17). This is clearly wrong; collisions cannot alter the free phonon ballistic propagation velocity at short times when few or no collisions occur.

At small $\omega \tau_{Q}$, the $\omega$-dependences of $n_{T}(\omega)$ and $d_{T}(\omega)$ are

$$\frac{1}{N} \sum_{r} r^{2} \Delta T(r,\omega) \to \frac{2}{\omega^{2}} \sum_{Q} \frac{\Delta E^\text{ext}_{Q}}{(1 + \omega \tau_{Q})^{2}} \sum_{Q} \frac{C_{Q} \Lambda_{Q}^{2}}{\tau_{Q}} + O\left(\frac{1}{\omega^{3}}\right), \quad (C18)$$

$$\frac{1}{N} \sum_{r} \Delta T(r,\omega) \to \frac{1}{\omega} \sum_{Q} \frac{\Delta E^\text{ext}_{Q}}{\tau_{Q}} \sum_{Q} \frac{C_{Q}}{\tau_{Q}} + O(1). \quad (C19)$$

The inverse Laplace transform of $2/\omega^{2}$ is $2t$, and of $1/\omega$ is 1. Then we get diffusive behavior in the long time limit, $\langle r^{2}\rangle_{T} \to 2Dt$, where $D = \kappa/C$

$$\kappa = \sum_{Q} C_{Q} \Lambda_{Q}^{2}/\tau_{Q} \quad \text{and} \quad C = \sum_{Q} C_{Q}. \quad (C20)$$

This result is independent of how the pulse inserts energy $\Delta E^\text{ext}_{Q}$, and the diffusivity $D$ has the correct macroscopic value. This contradicts the argument that pure diffusion isn’t in the nonlocal PBE.

Why would the nonlocal PBE give correct long time diffusion but incorrect short time ballistic? The answer, we think, is that version (1) of RTA, namely energy conservation $\sum_{Q} h\omega_{Q} \Phi_{Q} / \tau_{Q}$ doesn’t work when $\tau_{Q} \to \infty$. 

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