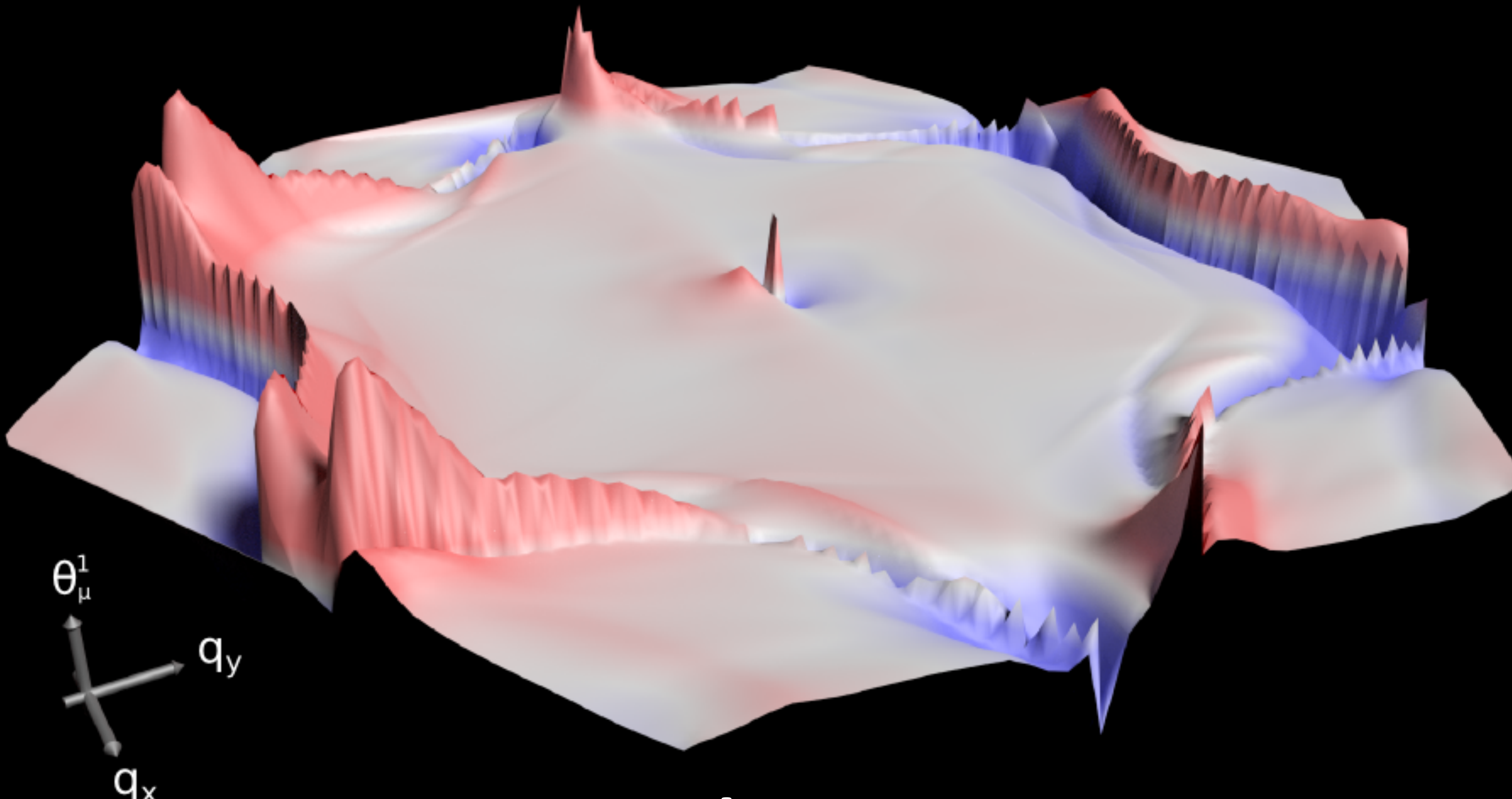


Emergent Phenomena in Phonon Thermal Transport

Berkeley
UNIVERSITY OF CALIFORNIA

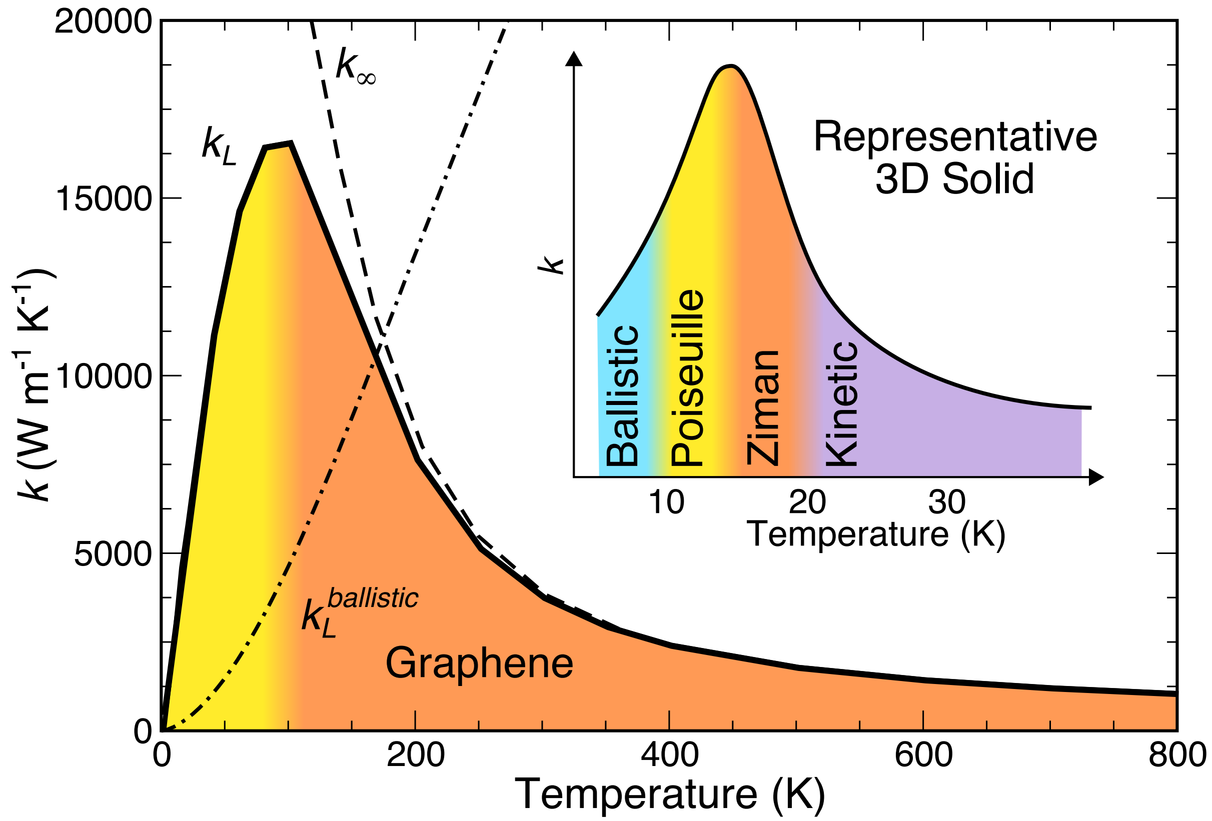
Andrea Cepellotti

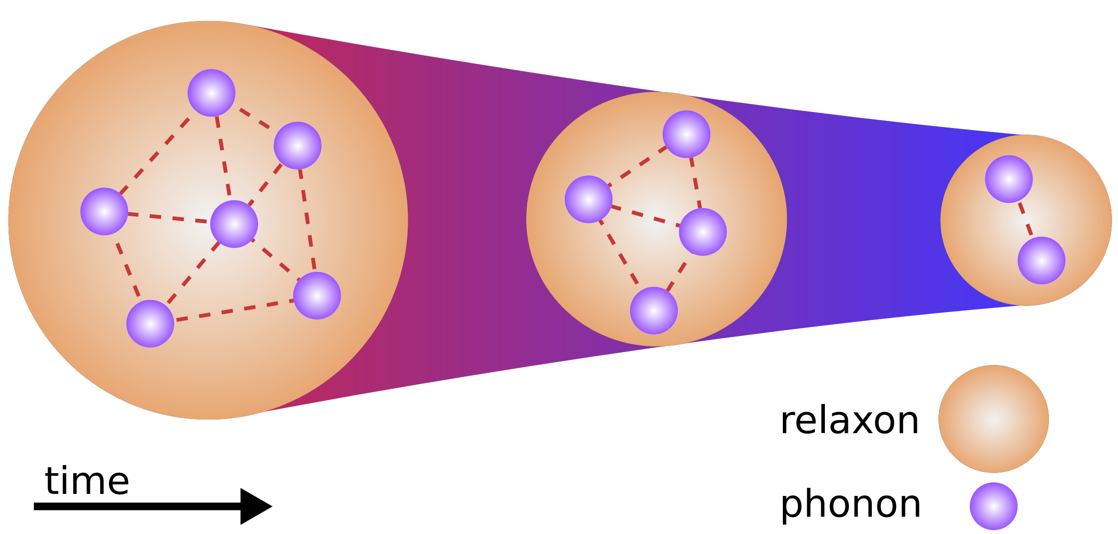
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

Lattice thermal transport

Outline

- Semiclassical Boltzmann transport equation;
- What is the relaxation time approximation and when does it fail;
- Formal definition of collective excitations: relaxons
- Surface scattering
- Second sound





Acknowledgements

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- Lorenzo Paulatto, UPMC, France
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- Steven G. Louie, UC Berkeley, USA

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MARVEL

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THEORY AND SIMULATION OF MATERIALS

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CSCS

Centro Svizzero di Calcolo Scientifico

Swiss National Supercomputing Centre

Lattice thermal transport

Phonon population at equilibrium: $\bar{n}_\mu = \dfrac{1}{e^{\hbar\omega_\mu / k_B T} - 1}$

$\mu = (\boldsymbol{q}, s)$
Index on all states

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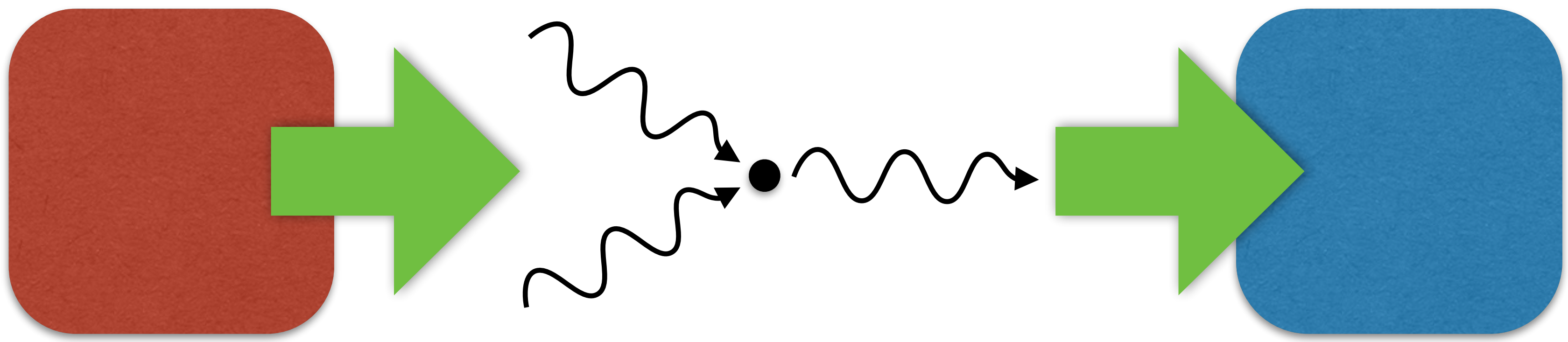

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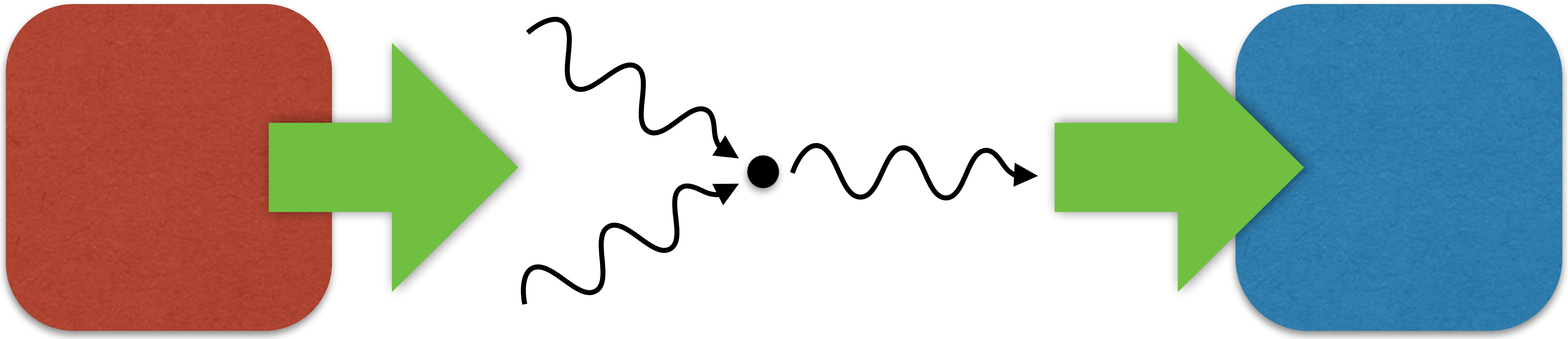

Linearised Boltzmann transport equation

$$\frac{\partial n_\mu}{\partial t} + \boldsymbol{v}_\mu \cdot \boldsymbol{\nabla} n_\mu = -\frac{1}{\mathcal{V}} \sum_{\mu'} \Omega_{\mu\mu'} \Delta n_{\mu'}$$

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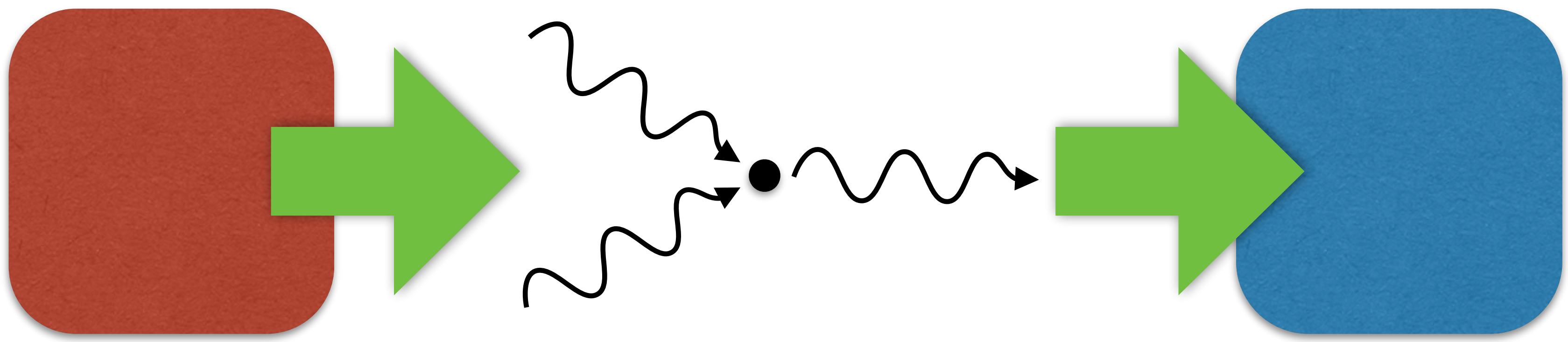

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Diffusion / Liouville operator

Collision / scattering operator (linearised)

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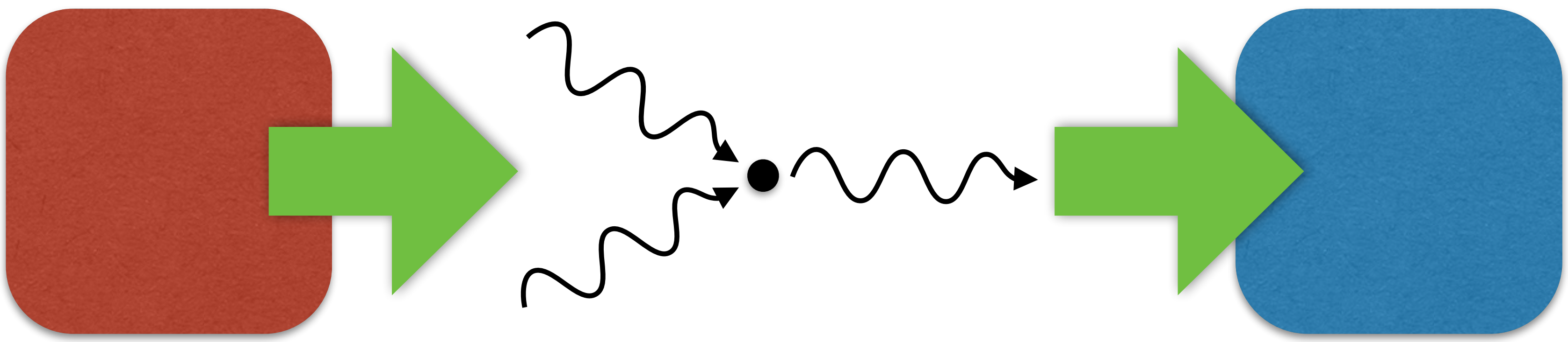

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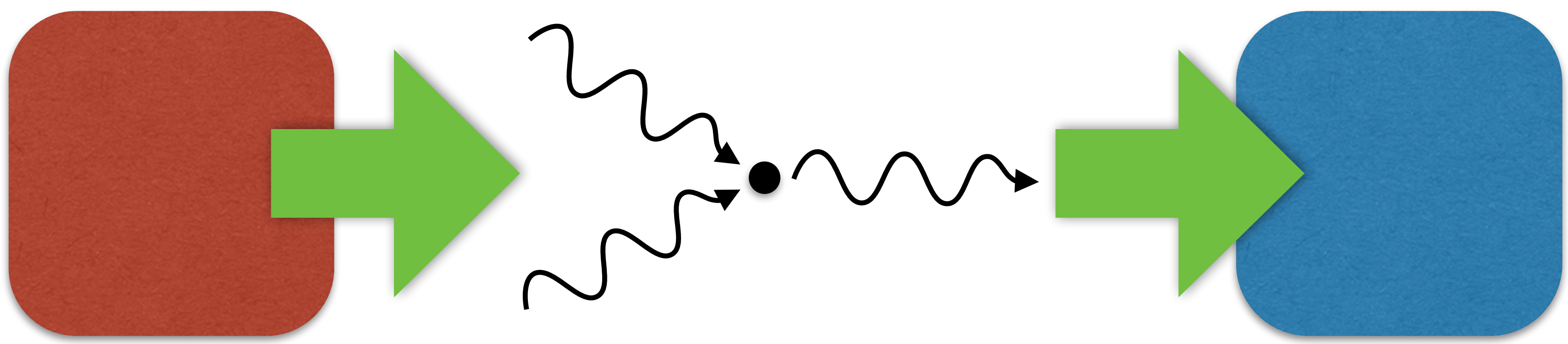

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Thermal conductivity: $k = -\dfrac{Q}{\nabla T} = -\dfrac{\sum_\mu \hbar\omega_\mu v_\mu \Delta n_\mu}{\mathcal{V}\nabla T}$

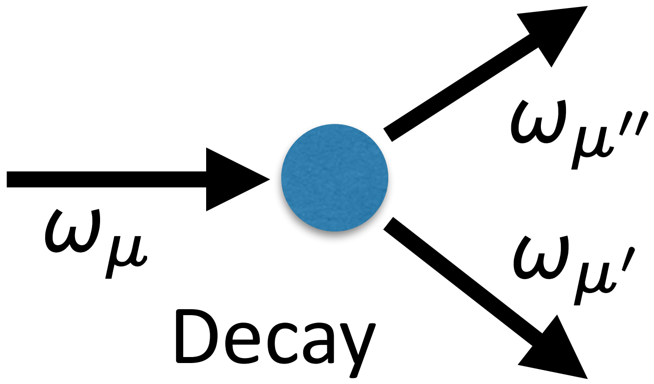
Within harmonic approximation (Hardy)

Boltzmann from first principles

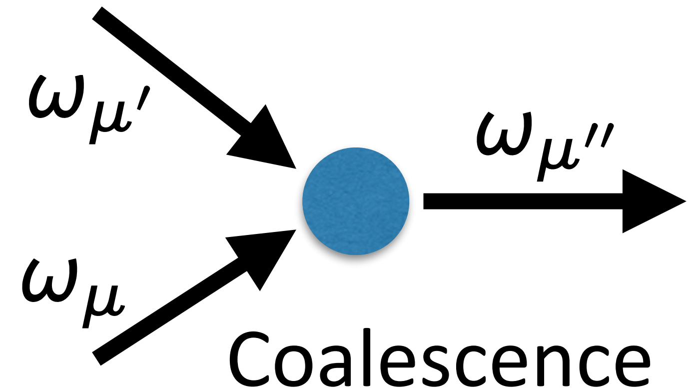
3-phonon interactions

Vanderbilt, Louie, Cohen PRB 33, 8740 (1986)

Vanderbilt, Taole, Narasimhan PRB 40, 5657 (1989)



Decay



Coalescence

Boltzmann from first principles

3-phonon interactions

Vanderbilt, Louie, Cohen PRB 33, 8740 (1986)

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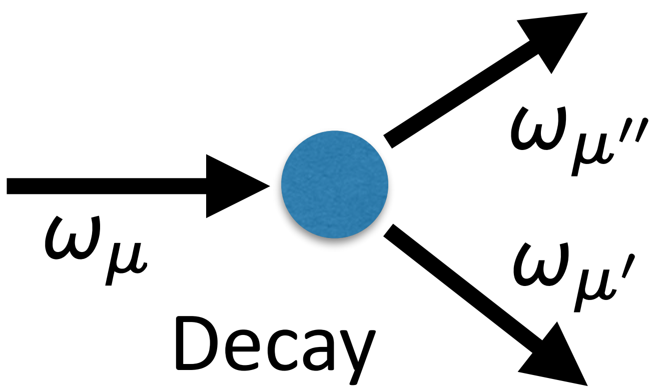
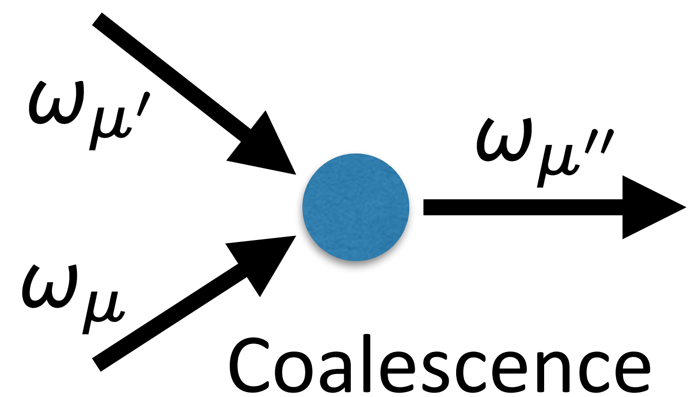
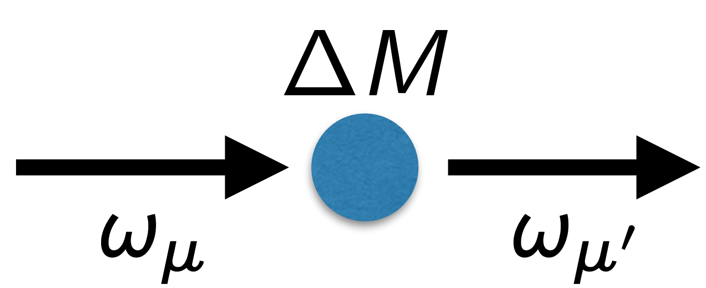




Mass disorder (isotopes)

Garg, Bonini, Kozinsky, Marzari PRL 106, 045901 (2011)



Boltzmann from first principles

Phonons properties with density functional perturbation theory

$$\frac{\partial^2 E}{\partial u_1 \partial u_2}$$

dynamical matrix
(provides phonon frequencies and eigenvectors)
Baroni, de Gironcoli, Dal Corso, Giannozzi
Rev. Mod. Phys. 75, 515 (2001)

$$\frac{\partial^3 E}{\partial u_1 \partial u_2 \partial u_3}$$

3rd-order anharmonic force constants
(provides 3-phonon coupling strengths)
Debernardi, Baroni, Molinari PRL 75, 1819 (1995)
Paulatto, Mauri, Lazzeri PRB 87, 214303 (2013)

Available in Quantum-ESPRESSO
www.quantum-espresso.org

Relaxation time approximation

Often, the Boltzmann equation is simplified using the Single-Mode relaxation time Approximation (SMA):

$$\frac{1}{\mathcal{V}}\sum_{\mu'}\Omega_{\mu\mu'}\Delta n_{\mu'} \approx \frac{\Delta n_{\mu}}{\tau_{\mu}^{\text{SMA}}} \longrightarrow$$

Time between phonon collisions

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C_μ: Phonon specific heat

v_μ: Phonon group velocity

$\Lambda_\mu^{\mathrm{SMA}}$: Phonon mean free path

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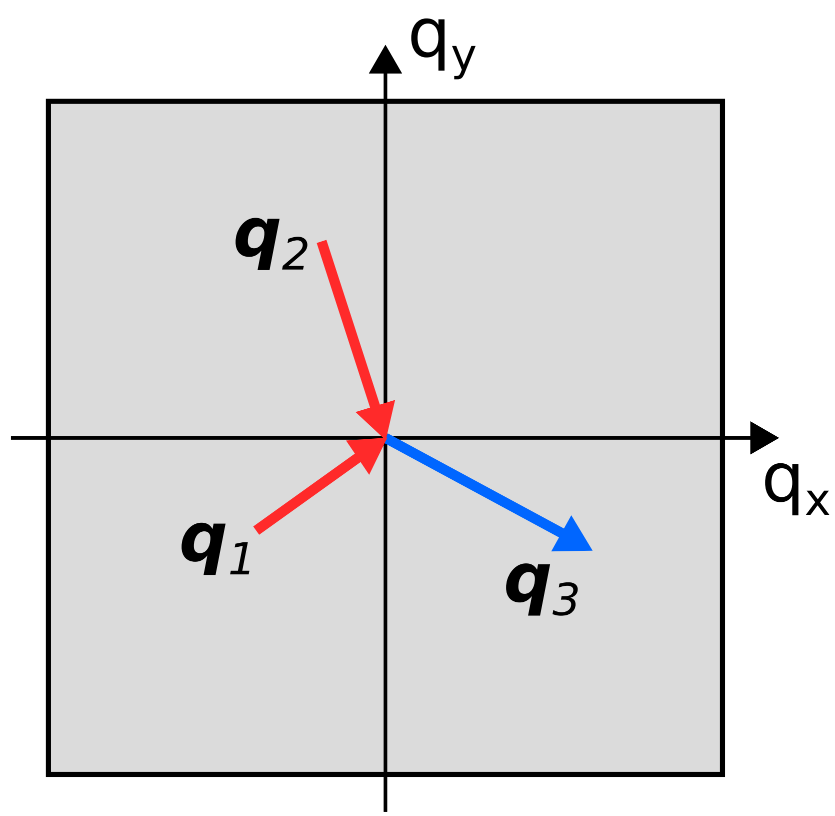
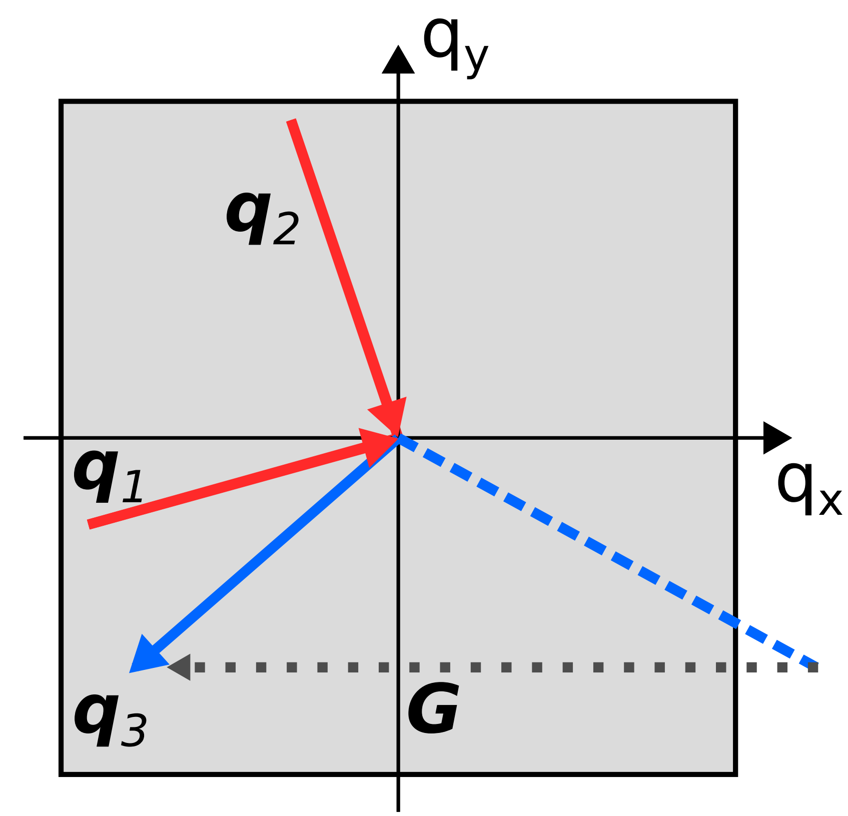
$$\Lambda_\mu^{\mathrm{SMA}} = v_\mu \tau_\mu^{\mathrm{SMA}}$$

$k \sim \tau_\mu^{\mathrm{SMA}} \Longrightarrow$ heat flux dissipated at every scattering event

What's wrong with the SMA?



Normal process

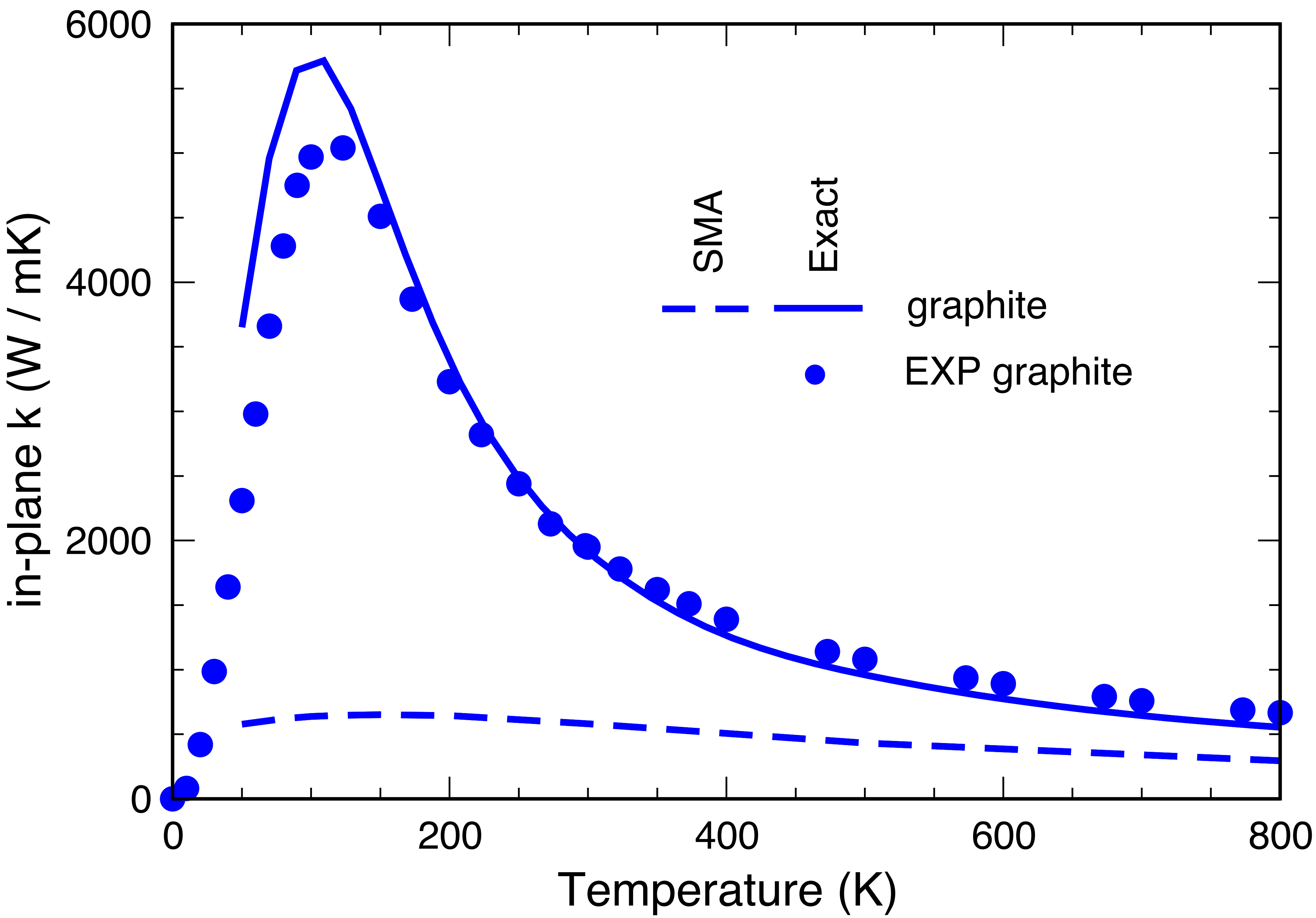


Umklapp process

	Normal	Umklapp
"Momentum" conservation	✔	✘
Heat flux conservation	✔	✘

Phonon scatterings don't always dissipate heat flux, as the SMA incorrectly assumes.

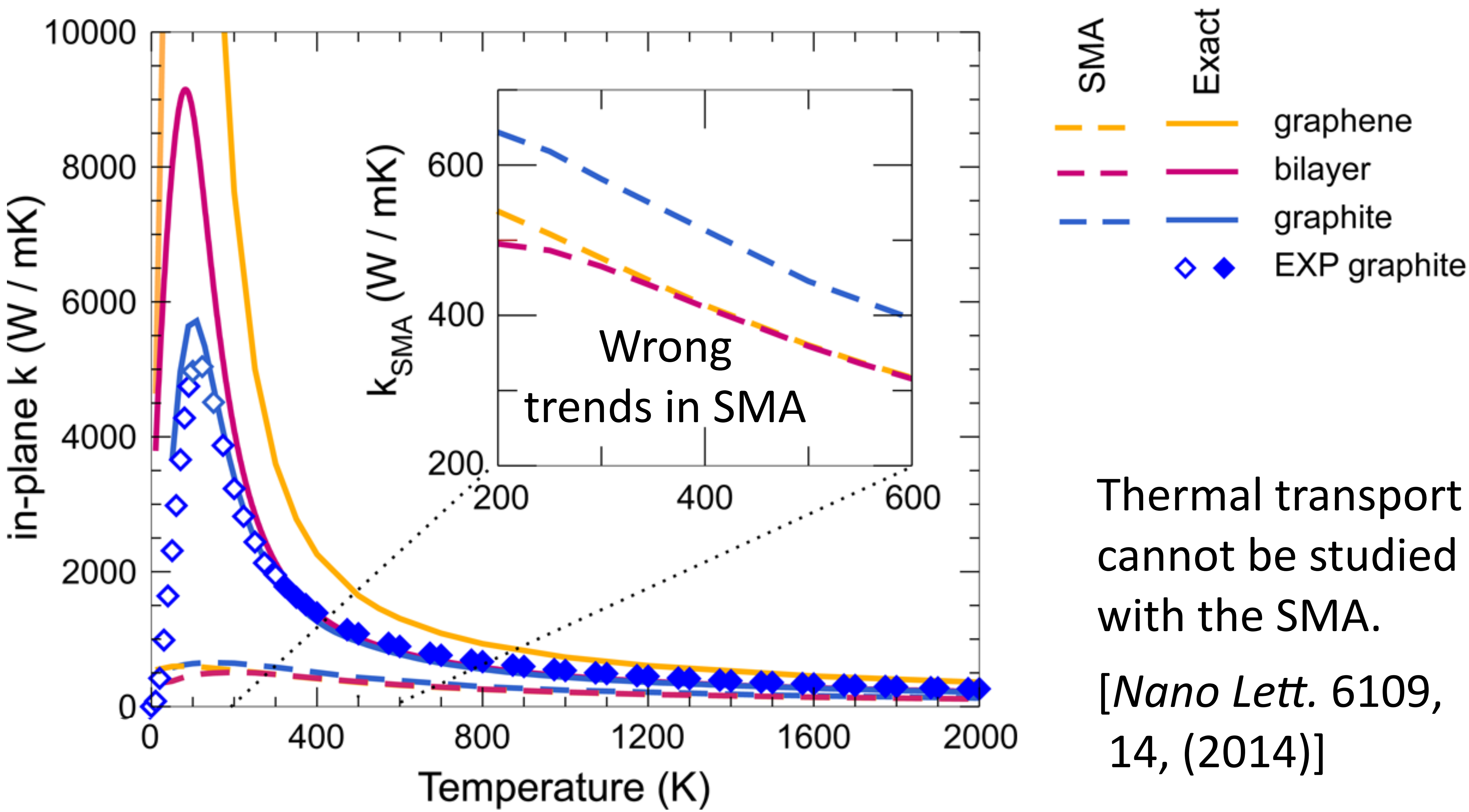
Graphite in-plane



The exact solution is necessary in graphite.

[*Nano Lett.* 6109, 14, (2014)]

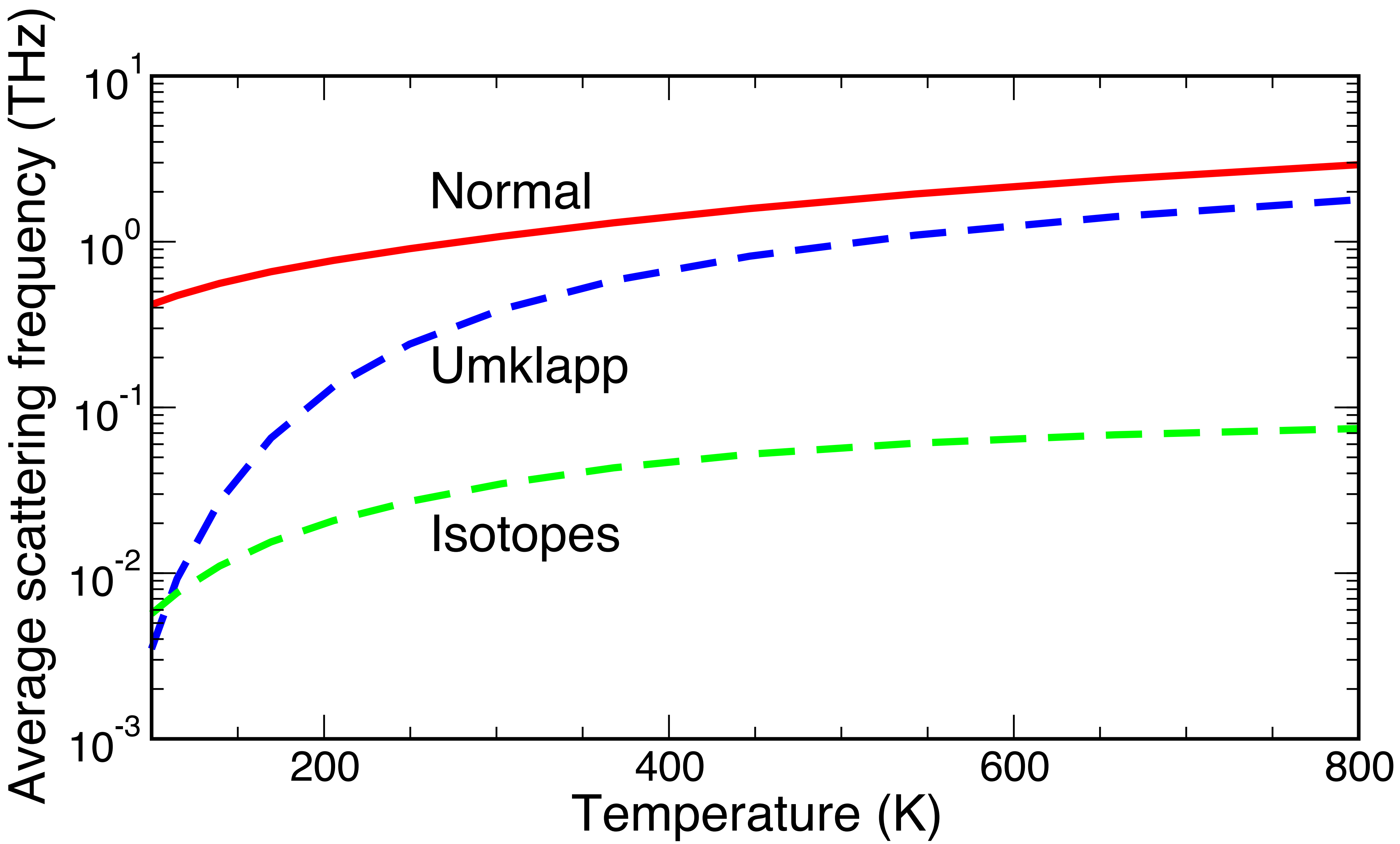
Graphite and graphene



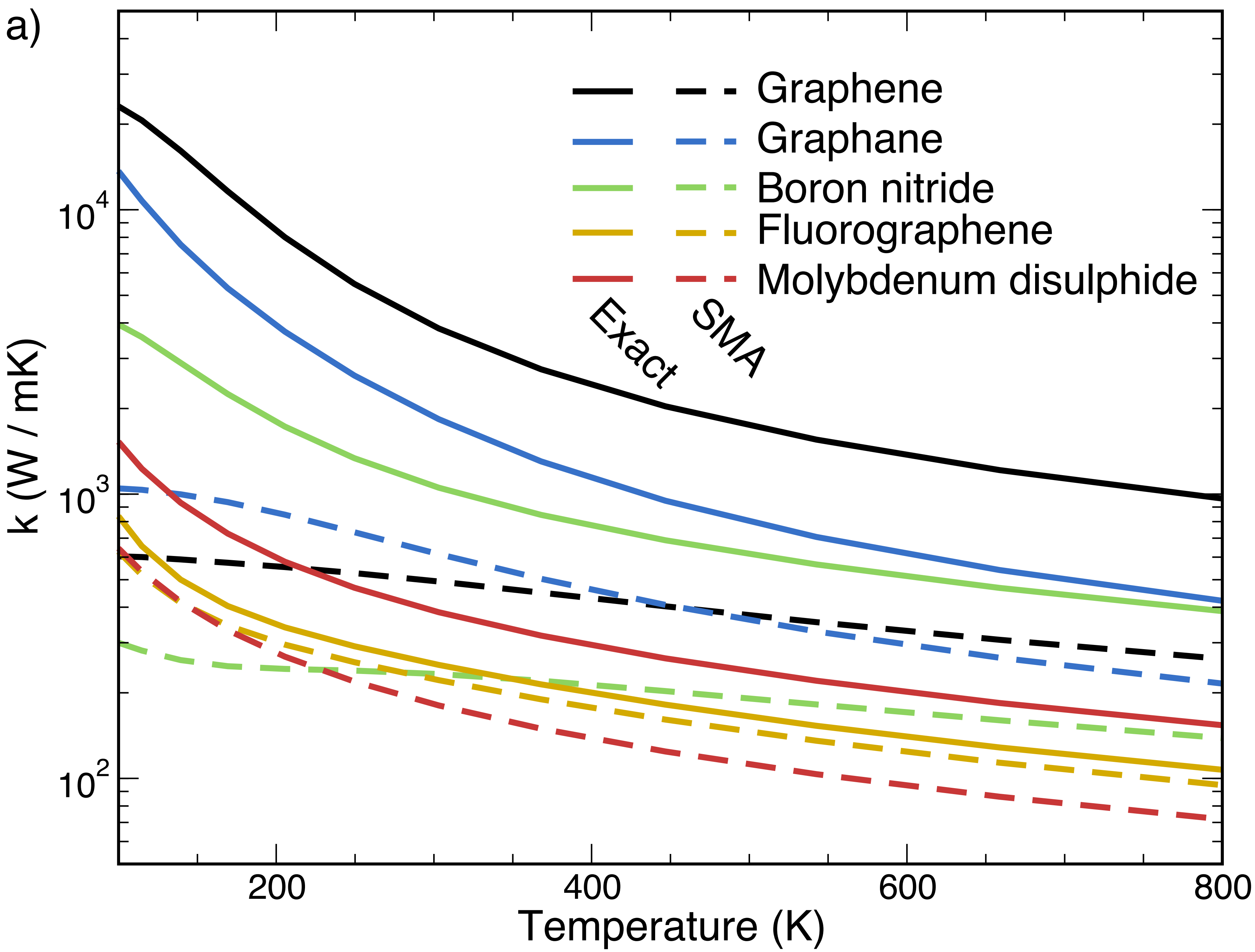
Thermal transport cannot be studied with the SMA.

[*Nano Lett.* 6109, 14, (2014)]

Graphene: Normal vs Umklapp



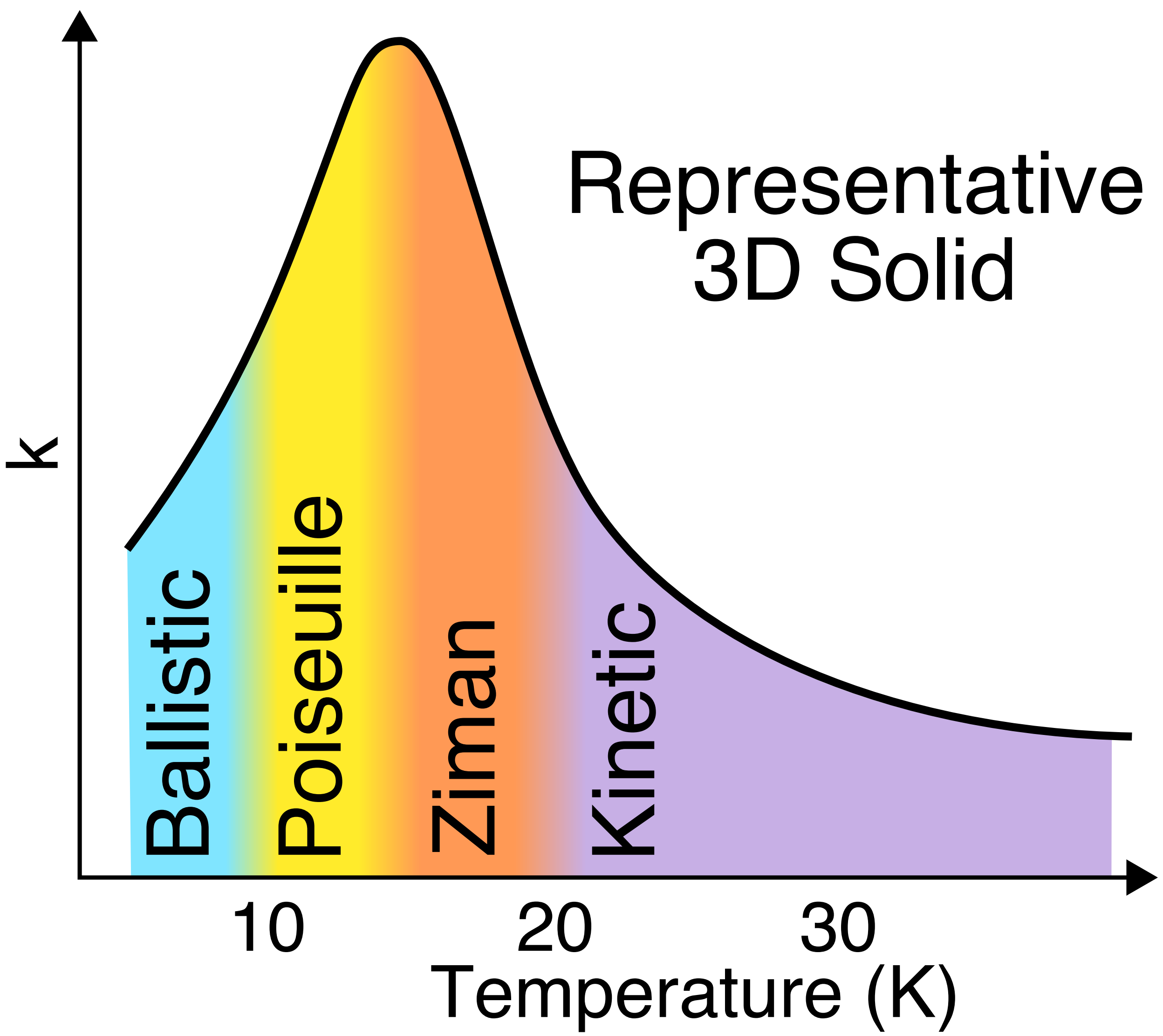
Exact vs SMA conductivity



The exact solution of the Boltzmann transport equation is necessary in 2D materials. [*Nat. Commun.* ***6***, *7400 (2015)*]

Failure first found in graphene: [*Lindsay et al. PRB* ***82***, *115427 (2010)]*

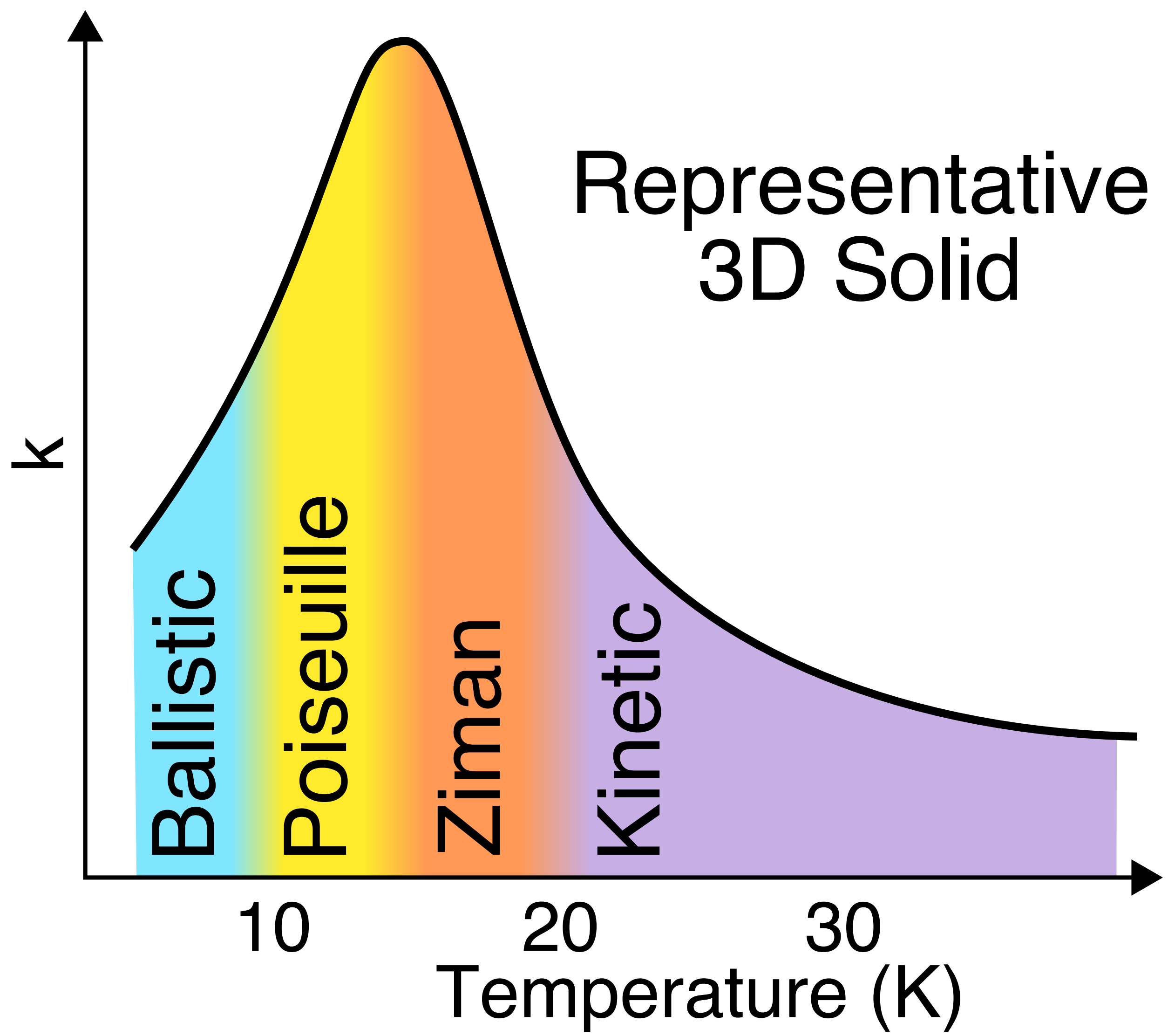
Heat transport regimes



Cepellotti A. et al.

Nat. Commun. ***6****, 7400 (2015)*

Heat transport regimes



Cepellotti A. et al.
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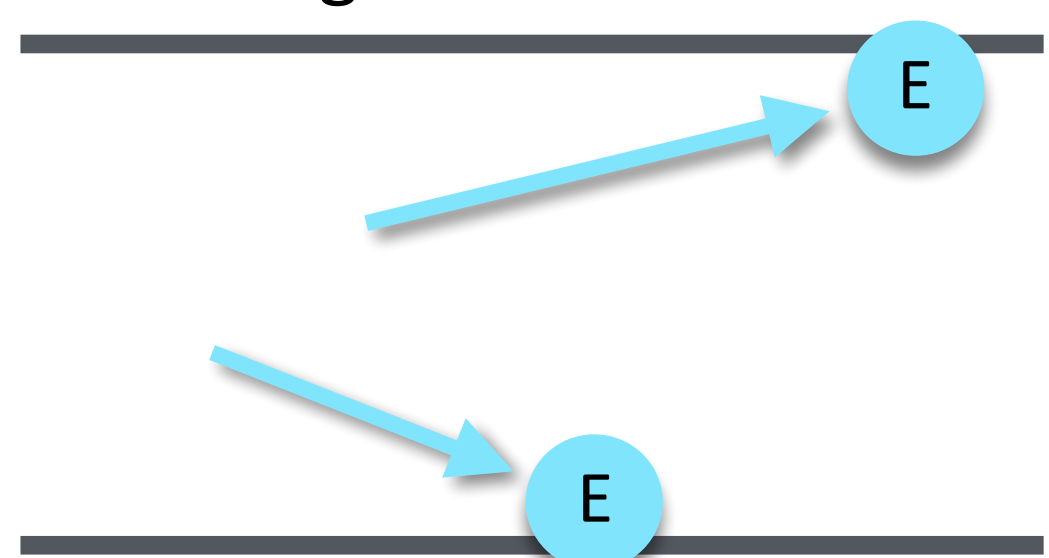
Ballistic	E≫N and E≫U

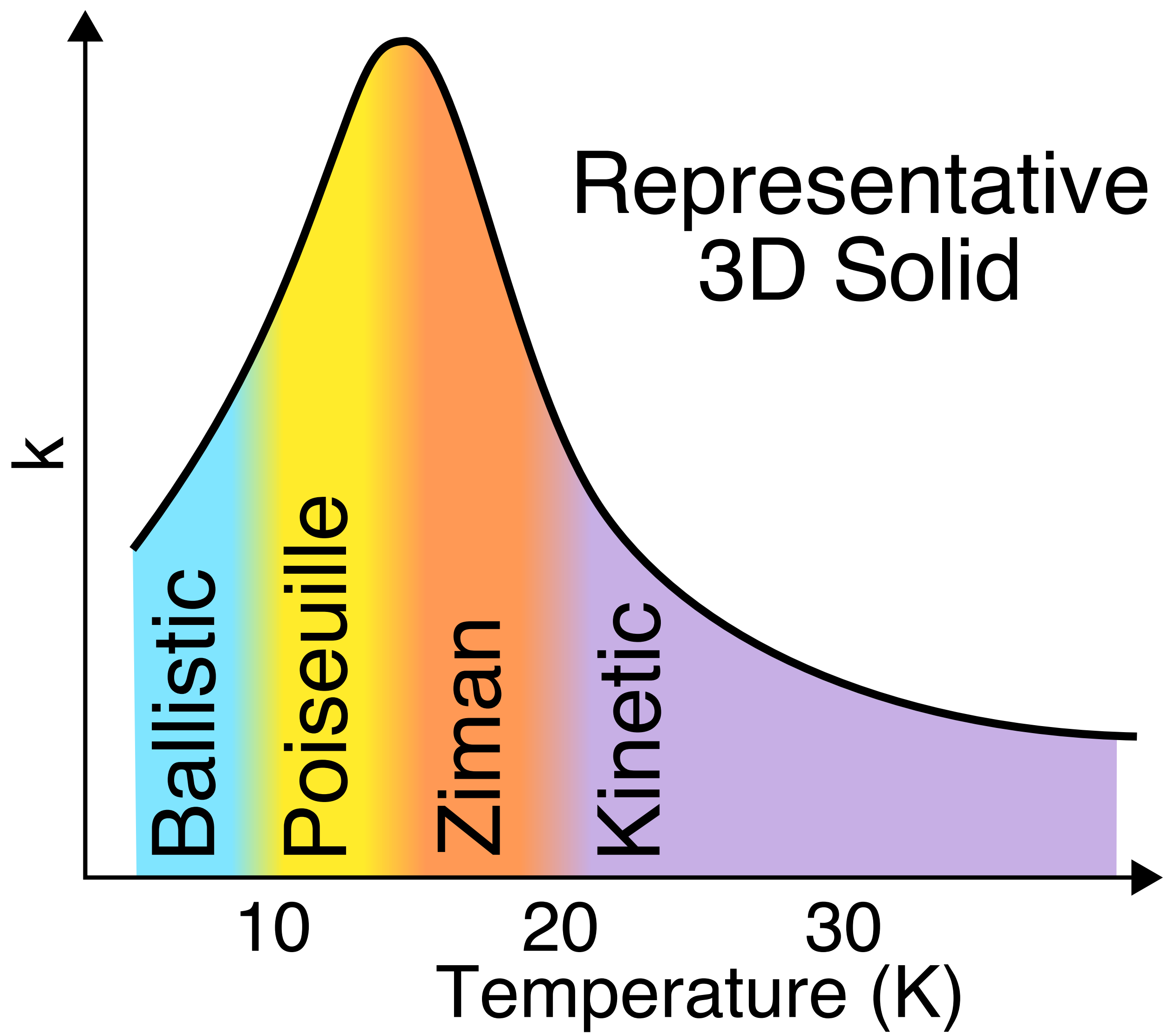
Scattering rates of extrinsic (E), Normal (N) and Umklapp (U) events

Ballistic regime:



Phonons scatter and dissipate heat only on the surfaces.

Heat transport regimes



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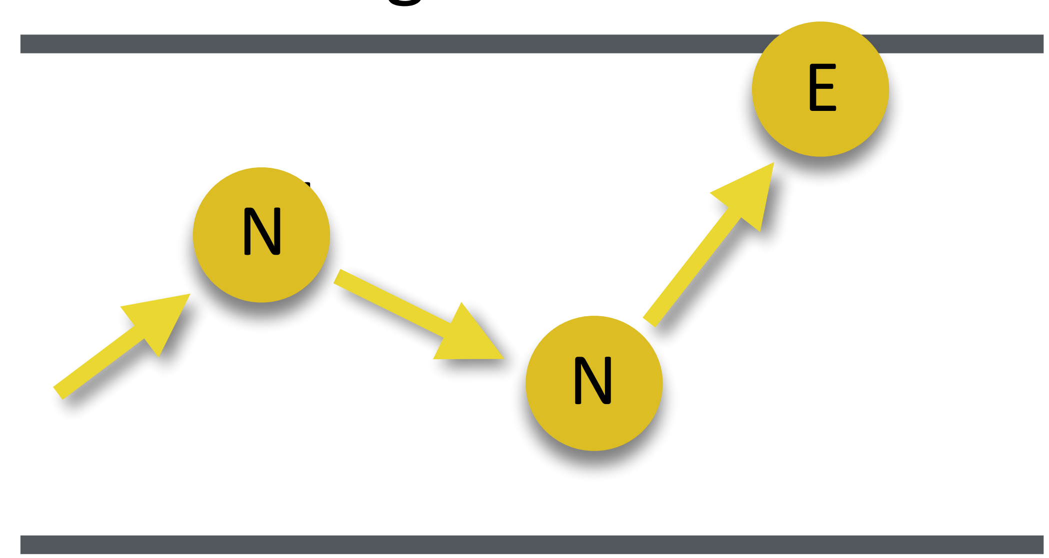
Poiseuille	N≫E≫U

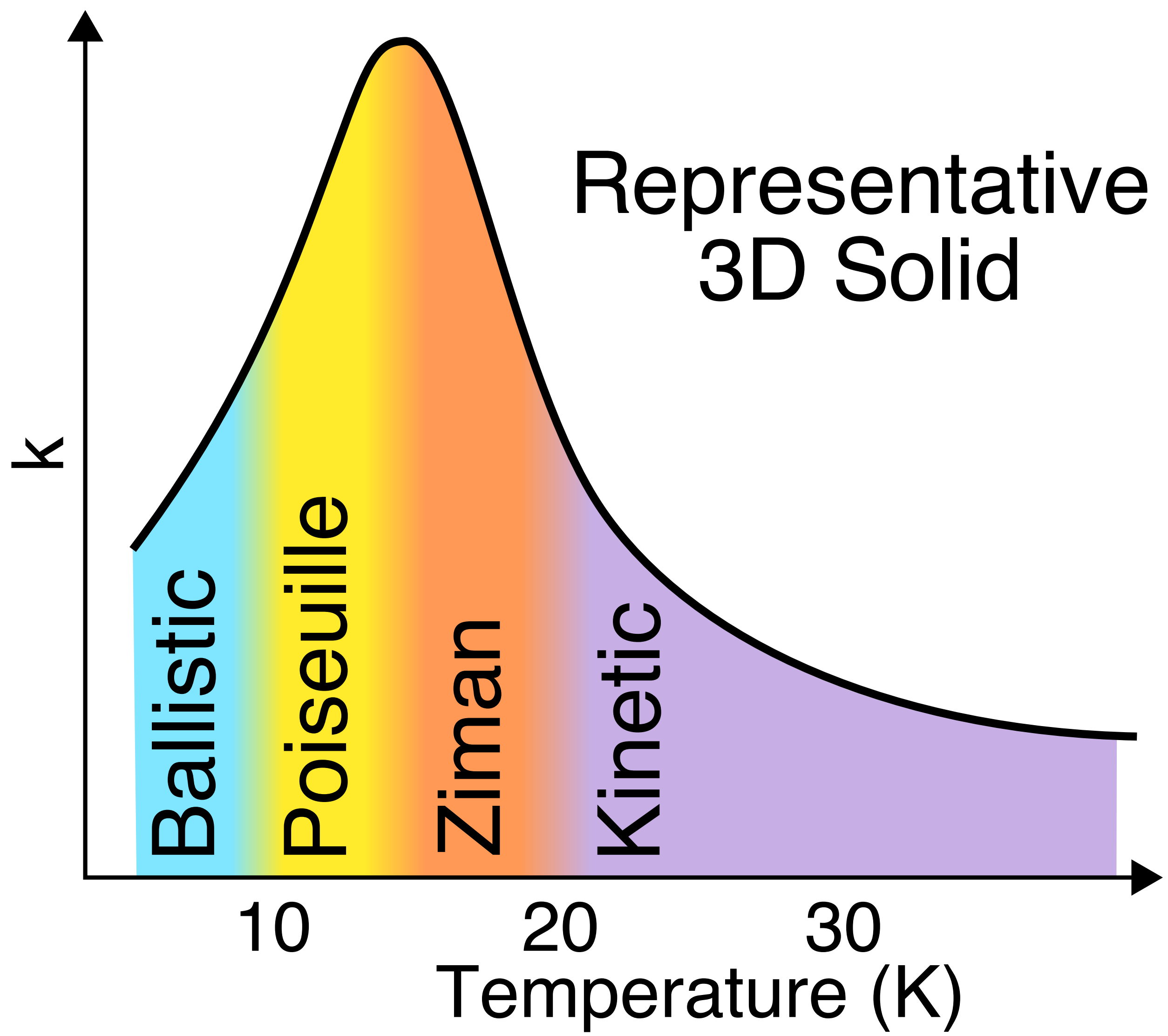
Scattering rates of extrinsic (E), Normal (N) and Umklapp (U) events

Poiseuille regime:



N processes dominate, and the phonon fluid still feels the "walls"

Heat transport regimes



Cepellotti A. et al.
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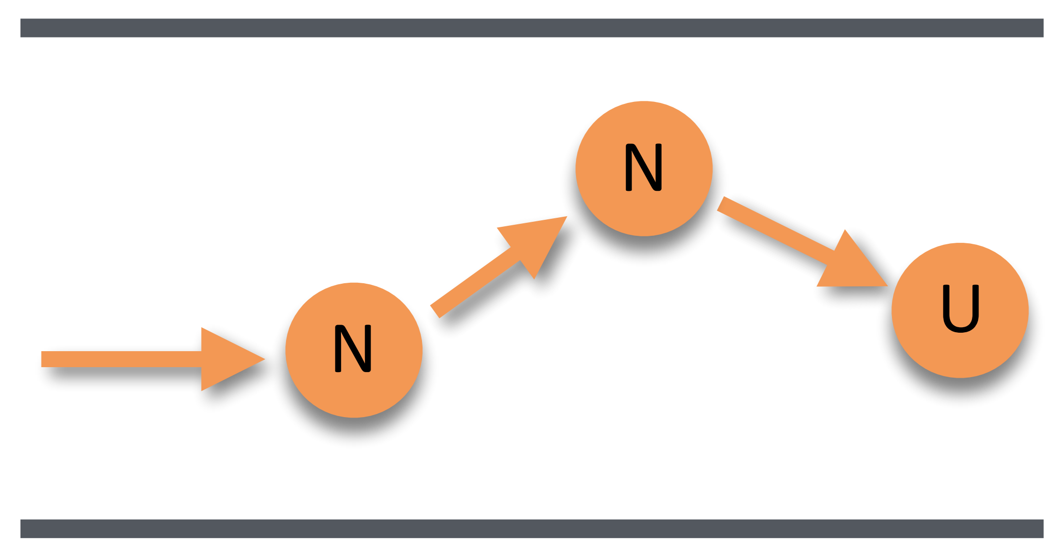
Ziman	N≫U≫E

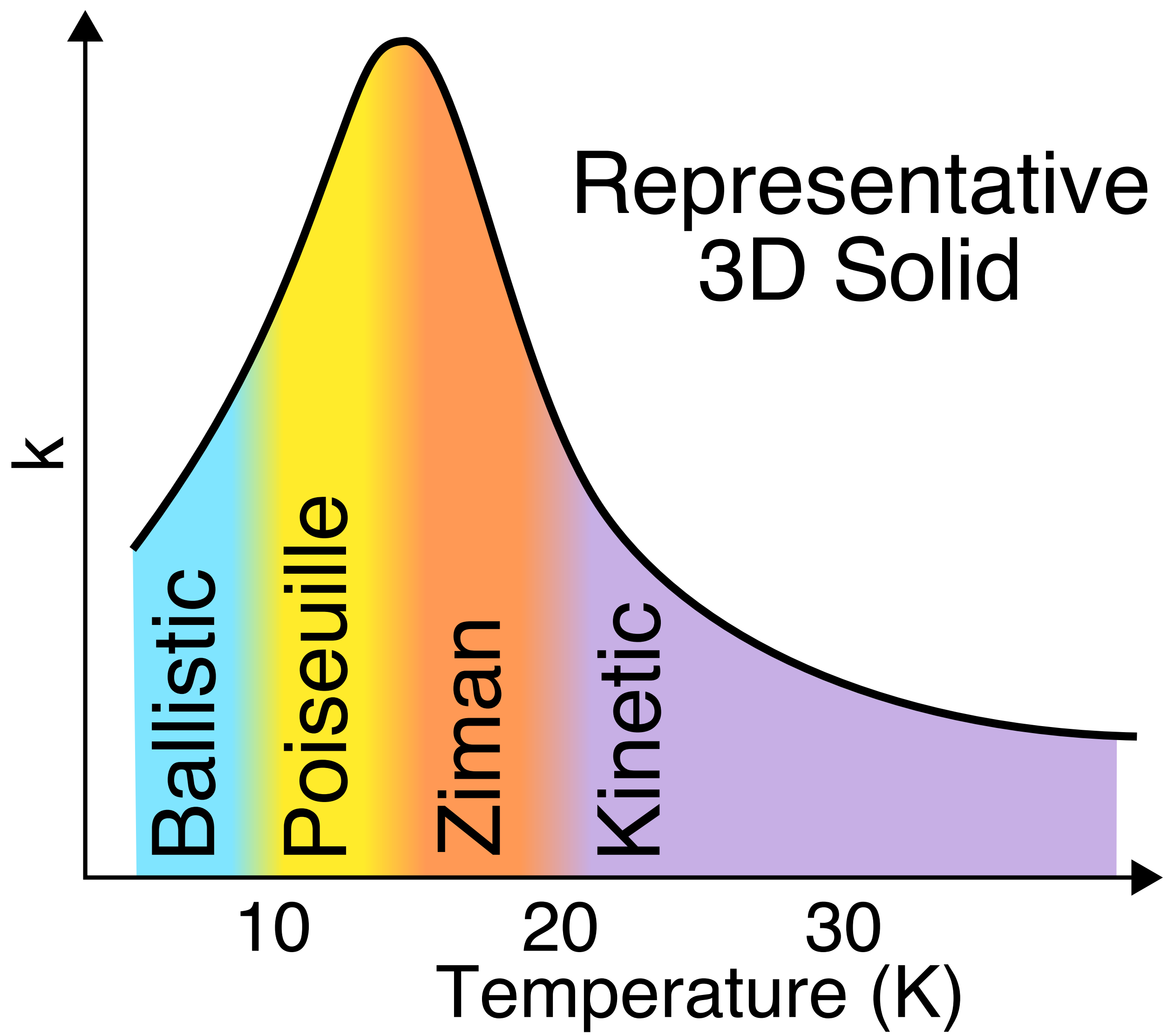
Scattering rates of extrinsic (E), Normal (N) and Umklapp (U) events

Ziman regime:



N processes dominate and heat is dissipated internally by U

Heat transport regimes



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Ballistic	E≫N and E≫U
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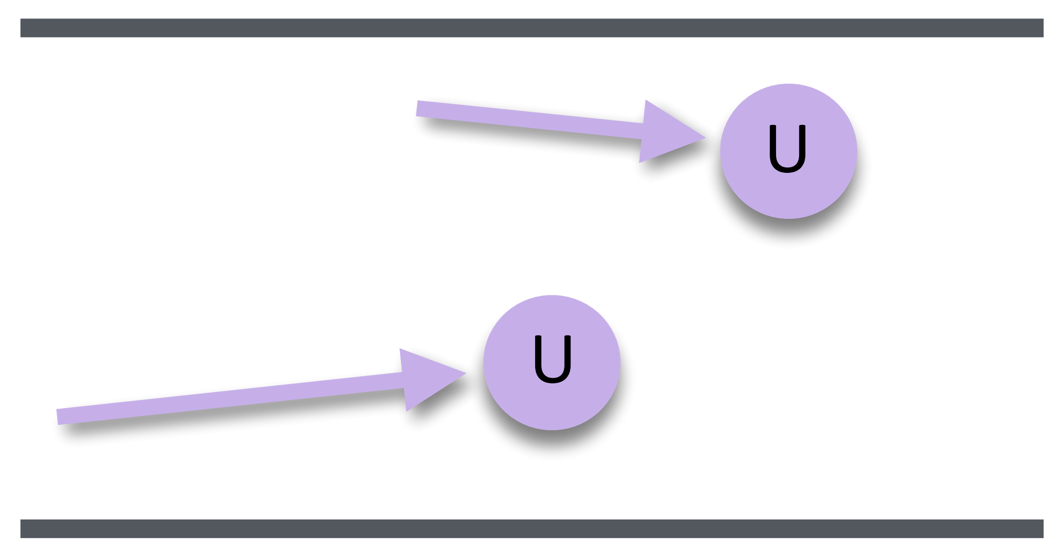
Ziman	N≫U≫E
Kinetic	U≫N and U≫E

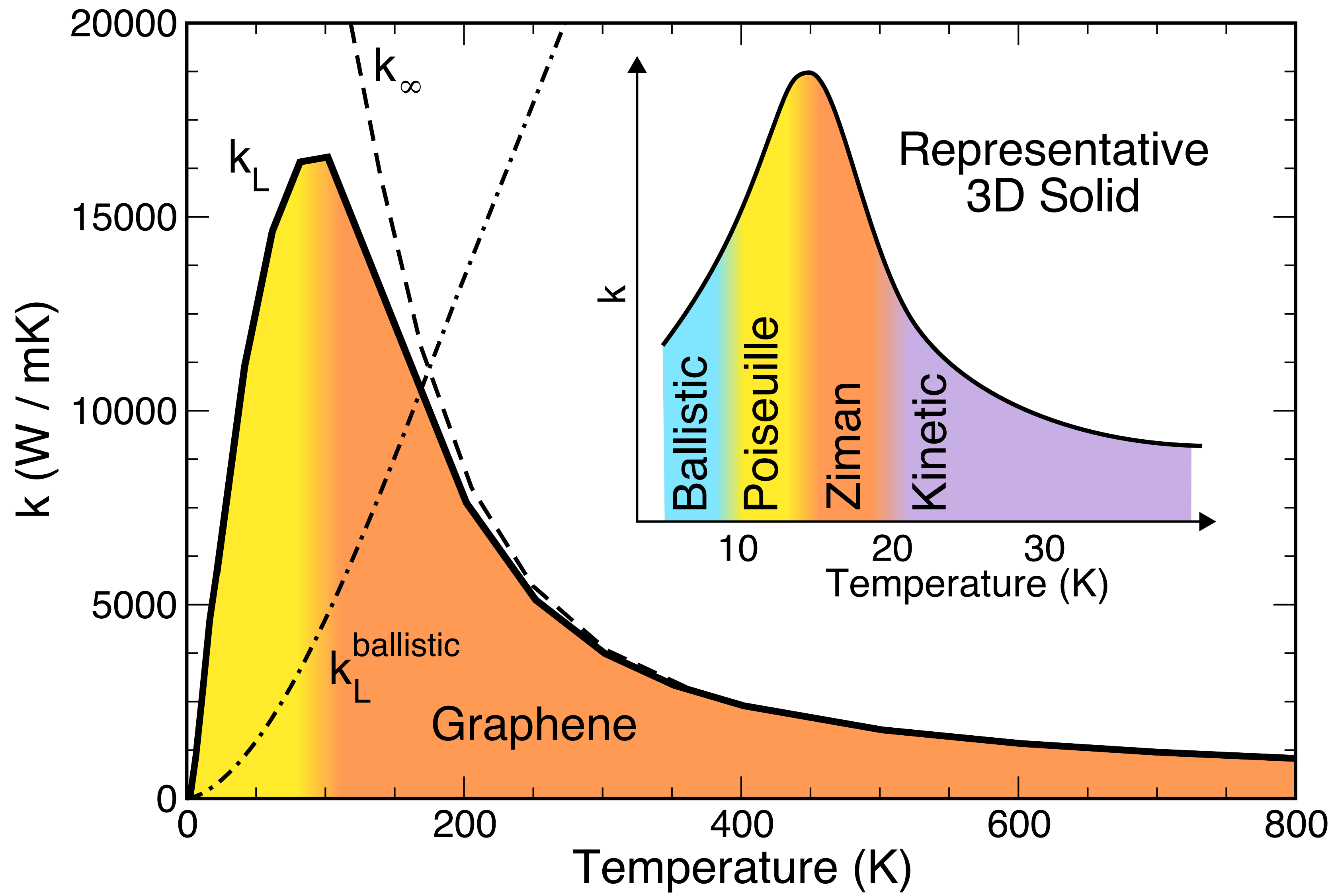
Scattering rates of extrinsic (E), Normal (N) and Umklapp (U) events

Kinetic regime:



The most common high-T regime where SMA (often) works

2D heat transport regimes



Ballistic	E≫N and E≫U
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Scattering rates of extrinsic (E), Normal (N) and Umklapp (U) processes

Hydrodynamic phonon transport is rarely present in 3D bulk systems.
In 2d materials, it's present at room temperature.

Cepellotti A. et al., Nat. Commun. ***6****, 7400 (2015)*

Defining heat carriers

Heat flux is not dissipated at every phonon scattering event.
Therefore phonons are not the heat carriers;

How can we define heat carriers?

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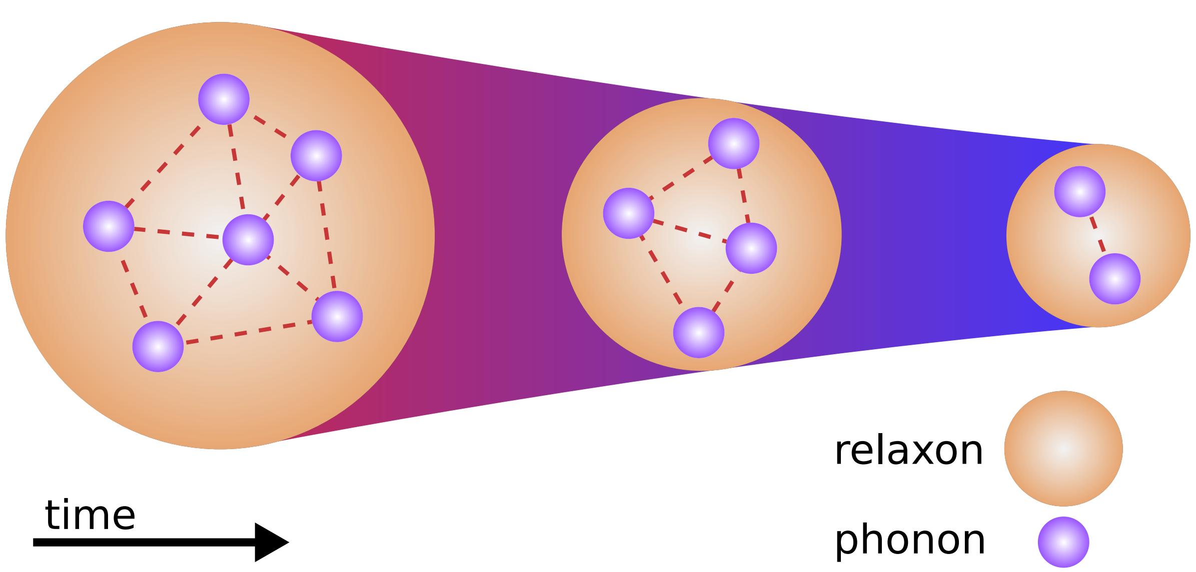
How can we define heat carriers?

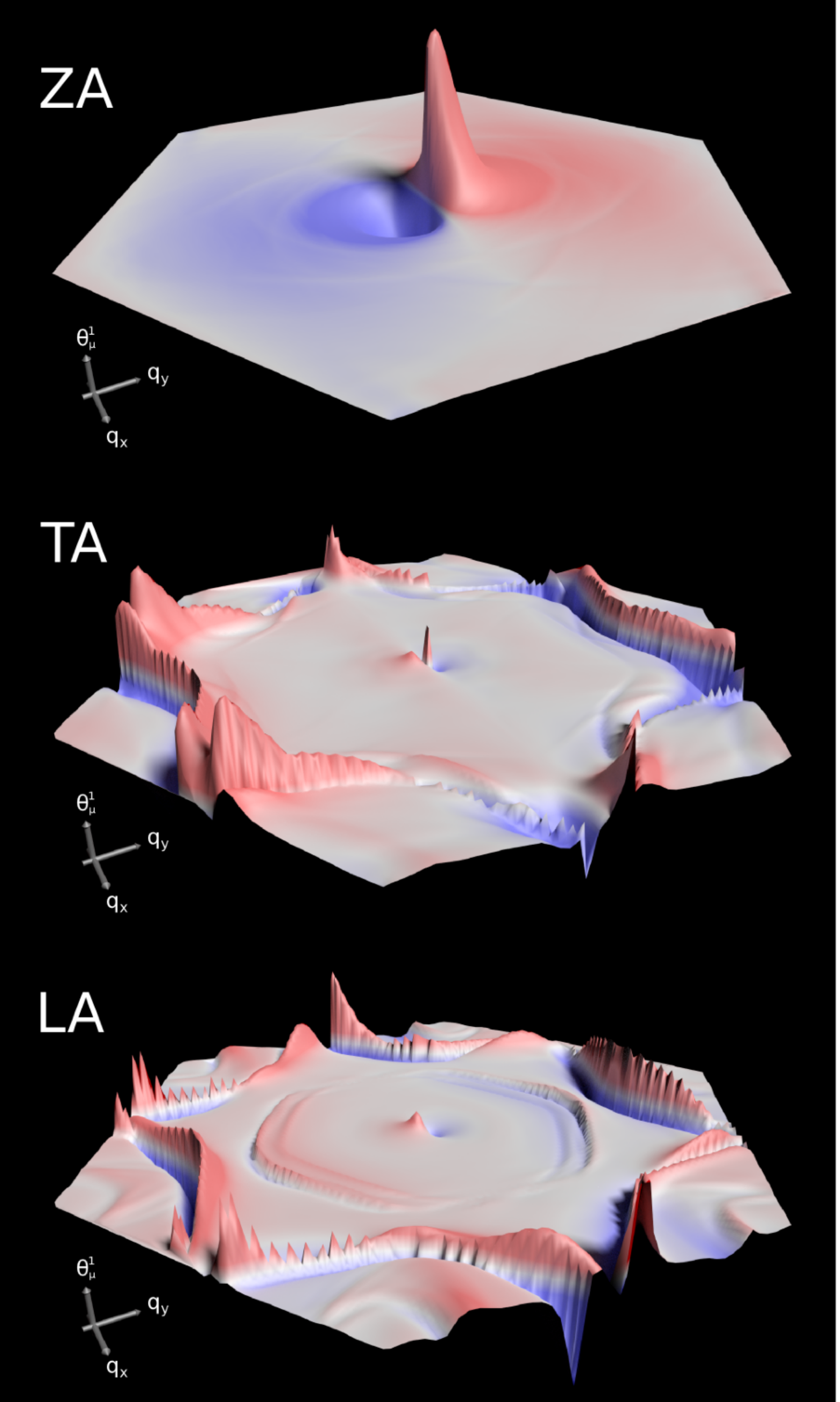
Our suggestion: **we diagonalise the scattering operator:**

$$\frac{1}{\mathcal{V}} \sum_{\mu'} \tilde{\Omega}_{\mu\mu'} \theta^{\alpha}_{\mu'} = \frac{1}{\tau_\alpha} \theta^{\alpha}_{\mu}$$

α labels all possible eigenvalues.

By definition, eigenvectors don't scatter among themselves.





Each eigenvector, termed relaxon, is a collective excitation of phonons.

Eigenvector = distribution of phonon populations

θ_μ^α

Eigenvalue index

Phonon index

Picture: eigenvector with smallest eigenvalue in graphene at 300K (ab-initio calculations)

Red areas indicate overpopulation of phonons w.r.t. thermal equilibrium, blue indicates depletion

From phonons to relaxons

Re-express phonon populations in terms of relaxon populations

$$\Delta n_\mu(\boldsymbol{x}, t) = \sum_\alpha f_\alpha(\boldsymbol{x}, t)\theta_\mu^\alpha$$

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Change basis of the Boltzmann transport equation

$$\sqrt{\frac{C}{k_B T^2}}\left(\frac{\partial T(\boldsymbol{x}, t)}{\partial t}\langle 0|\alpha\rangle + \nabla T(\boldsymbol{x}, t)\cdot \boldsymbol{V}_\alpha\right) + \frac{\partial f_\alpha(\boldsymbol{x}, t)}{\partial t} + \sum_{\alpha'} \boldsymbol{V}_{\alpha\alpha'}\cdot\nabla f_{\alpha'}(\boldsymbol{x}, t) = -\frac{f_\alpha(\boldsymbol{x}, t)}{\tau_\alpha}$$

Instead of phonons, we study relaxon populations f_α.

Exact relaxation times

Consider a system at thermal equilibrium where only one mode is excited uniformly in space (T=const and $\nabla f=0$):

$$\sqrt{\frac{C}{k_B T^2}}\left(\frac{\partial T(\boldsymbol{x},t)}{\partial t}\langle 0|\alpha\rangle + \nabla T(\boldsymbol{x},t)\cdot \boldsymbol{V}_\alpha\right) + \frac{\partial f_\alpha(\boldsymbol{x},t)}{\partial t} + \sum_{\alpha'} \boldsymbol{V}_{\alpha\alpha'}\cdot\nabla f_{\alpha'}(\boldsymbol{x},t) = -\frac{f_\alpha(\boldsymbol{x},t)}{\tau_\alpha}$$

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Populating a state α at time t_0, it relaxes as an exponential:

$$f_\alpha(t) = f_\alpha(t_0) e^{-t/\tau_\alpha}$$

Relaxons have a relaxation time!

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Relaxons have a relaxation time!

Rotating back to phonons: $$\Delta n_\mu(t) = \sum_\alpha \theta_\mu^\alpha f_\alpha(t_0) e^{-t/\tau_\alpha}$$

[PNAS, 113, 43 (2016)]

The phonon decay depends on the initial conditions:

⇒ ill-defined phonon relaxation times

Only relaxons have well-defined relaxation times!

Bulk thermal conductivity

Consider steady state (time disappears) and a bulk system (no spatial depencence):

$$\sqrt{\frac{C}{k_B T^2}} \left(\frac{\partial T(\boldsymbol{x}, t)}{\partial t} \langle 0 | \alpha \rangle + \nabla T(\boldsymbol{x}, t) \cdot \boldsymbol{V}_{\alpha} \right) + \frac{\partial f_{\alpha}(\boldsymbol{x}, t)}{\partial t} + \sum_{\alpha'} \boldsymbol{V}_{\alpha\alpha'} \cdot \nabla f_{\alpha'}(\boldsymbol{x}, t) = -\frac{f_{\alpha}(\boldsymbol{x}, t)}{\tau_{\alpha}}$$

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$$\sqrt{\frac{C}{k_B T^2}}V_\alpha^i = -\frac{f_\alpha^i}{\tau_\alpha}$$

Which we can solve analytically!

Bulk thermal conductivity

The thermal conductivity in the basis of relaxons is:

$$k = \sum_{\alpha} C V_{\alpha} \Lambda_{\alpha}$$

Bulk thermal conductivity

The thermal conductivity in the basis of relaxons is:

$$k = \sum_{\alpha} C V_{\alpha} \Lambda_{\alpha}$$

$$\Lambda_{\alpha} = V_{\alpha} \tau_{\alpha}$$

Relaxon mean free path

Relaxon specific heat

C, same ∀ relaxon

Relaxon velocity

$$\boldsymbol{V}_{\alpha} = \frac{1}{\mathcal{V}} \sum_{\mu} \theta_{\mu}^{0} \boldsymbol{v}_{\mu} \theta_{\mu}^{\alpha}$$

Bulk thermal conductivity

The thermal conductivity in the basis of relaxons is:

$$k = \sum_{\alpha} C V_\alpha \Lambda_\alpha$$

$$\Lambda_\alpha = V_\alpha \tau_\alpha$$

Relaxon mean free path

Relaxon specific heat

C, same $\forall$ relaxon

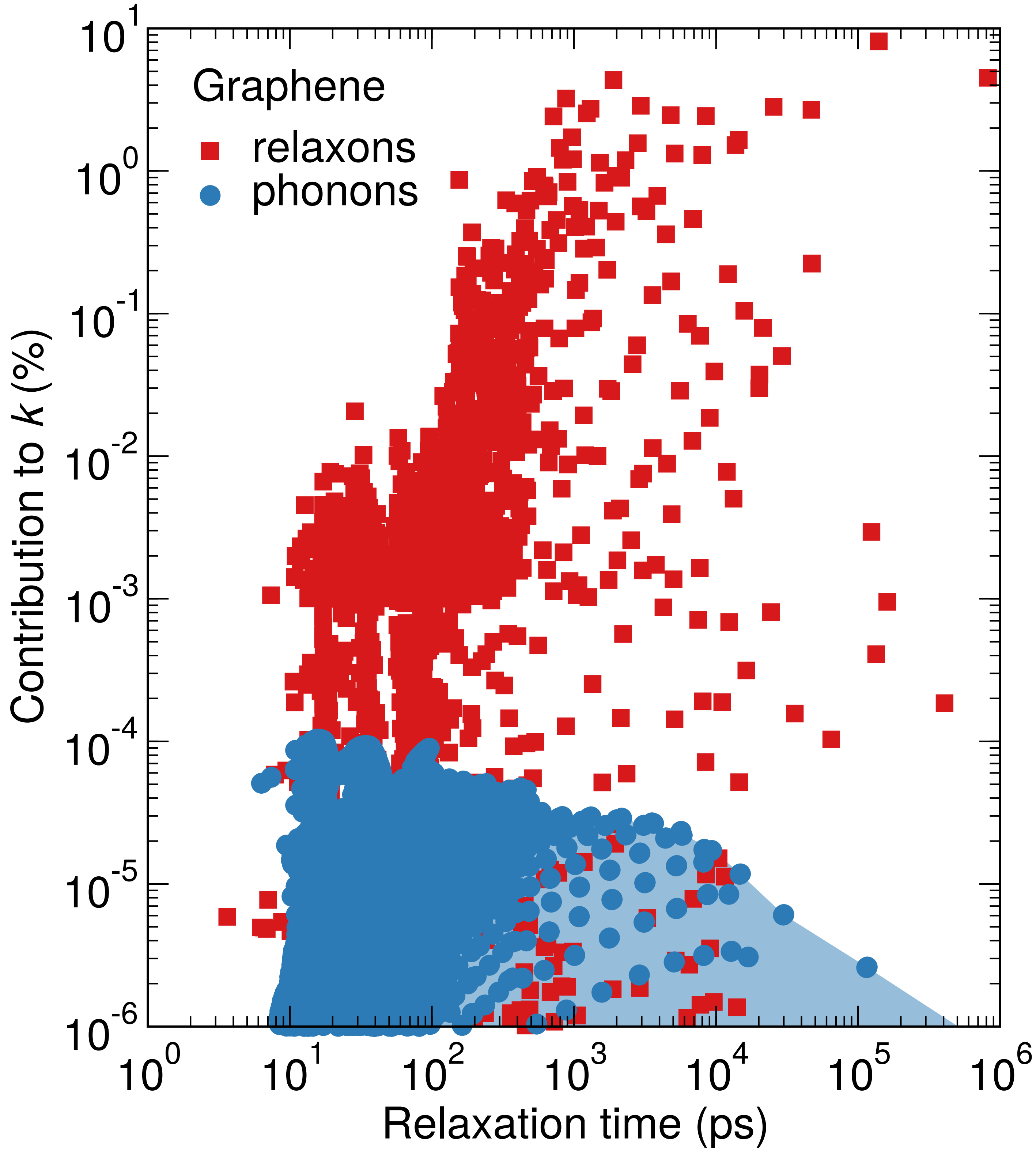
Relaxon velocity

$$\boldsymbol{V}_\alpha = \frac{1}{\mathcal{V}} \sum_{\mu} \theta^0_\mu \boldsymbol{v}_\mu \theta^\alpha_\mu \qquad \theta^0_\mu = \frac{\sqrt{\bar{n}_\mu(\bar{n}_\mu + 1)\hbar\omega_\mu}}{\sqrt{C k_B T^2}}$$

We recover a kinetic-gas like description of thermal transport, with new estimates of time, length and velocity scales of transport.

$\Longrightarrow$ we identify relaxons with the heat carriers

Graphene @ 300K



Phonon lifetimes

vs

Relaxon relaxation times.

Phonon scattering time scale

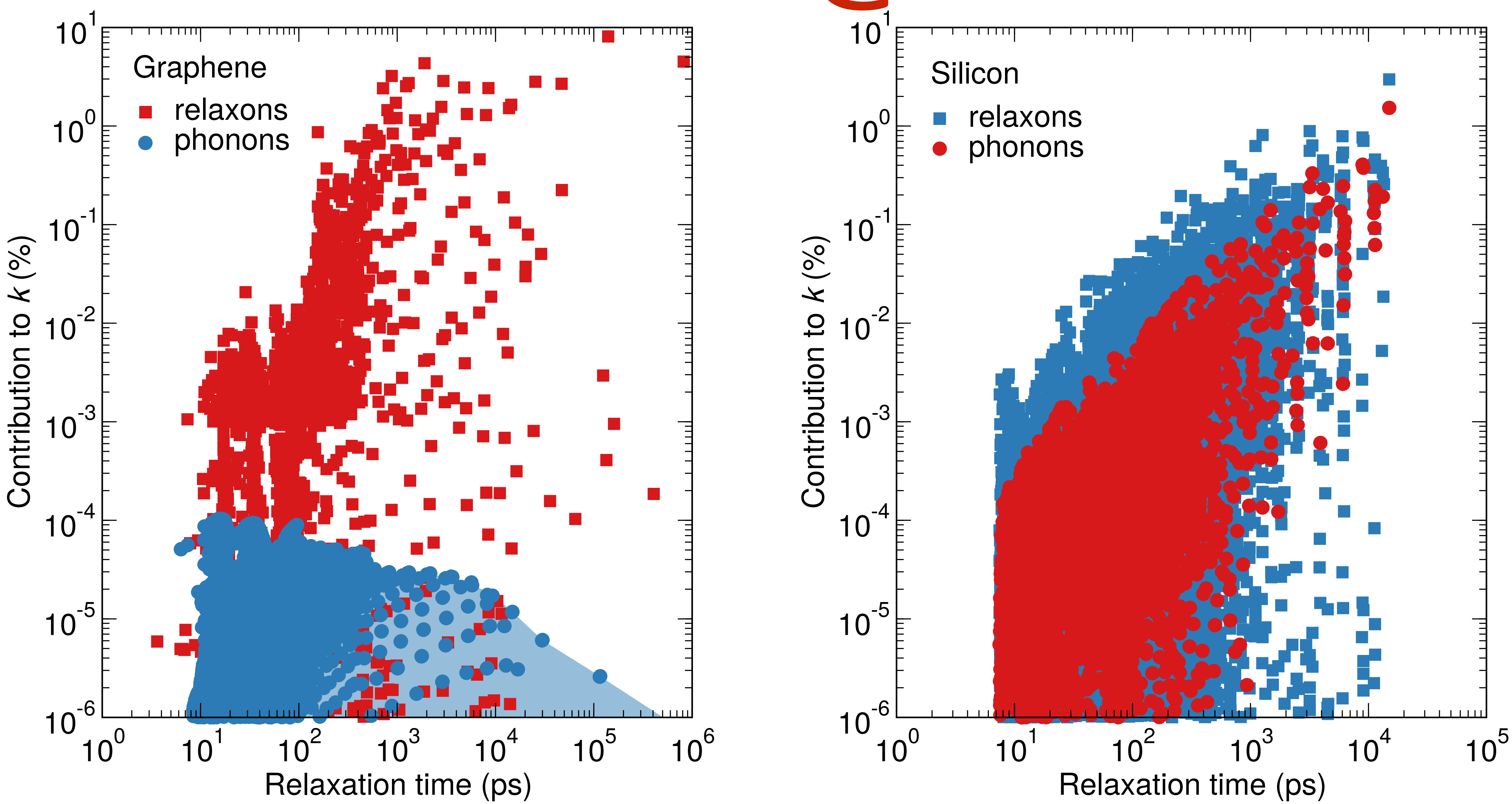
≈ 10-100ps

Heat flux time scale

>> 1000ps

[PRX 6, 041013 (2016)]

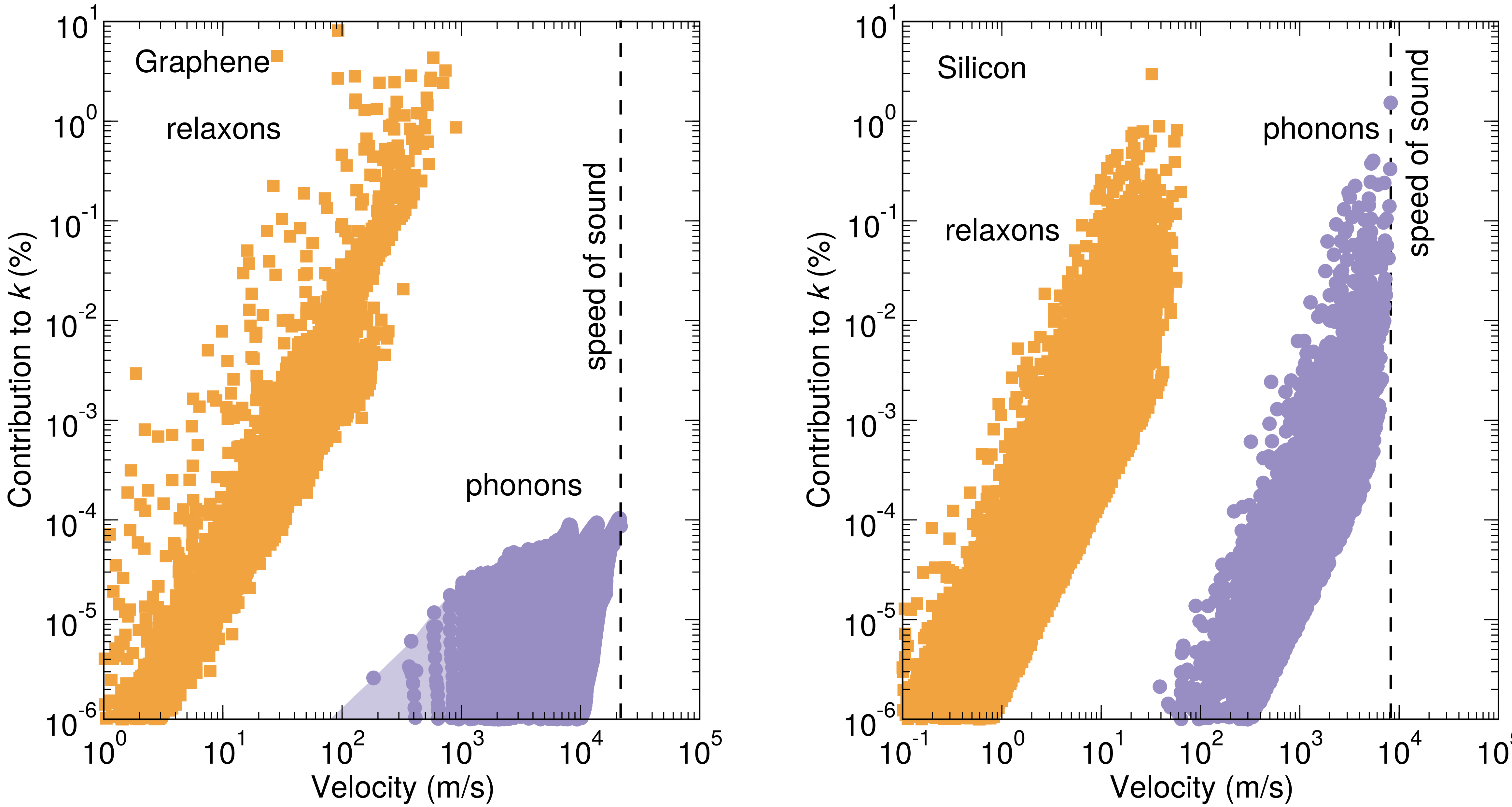
Lifetimes @ 300K



Two examples: graphene (where the SMA underestimates fails) and silicon (where the SMA gives good conductivity)

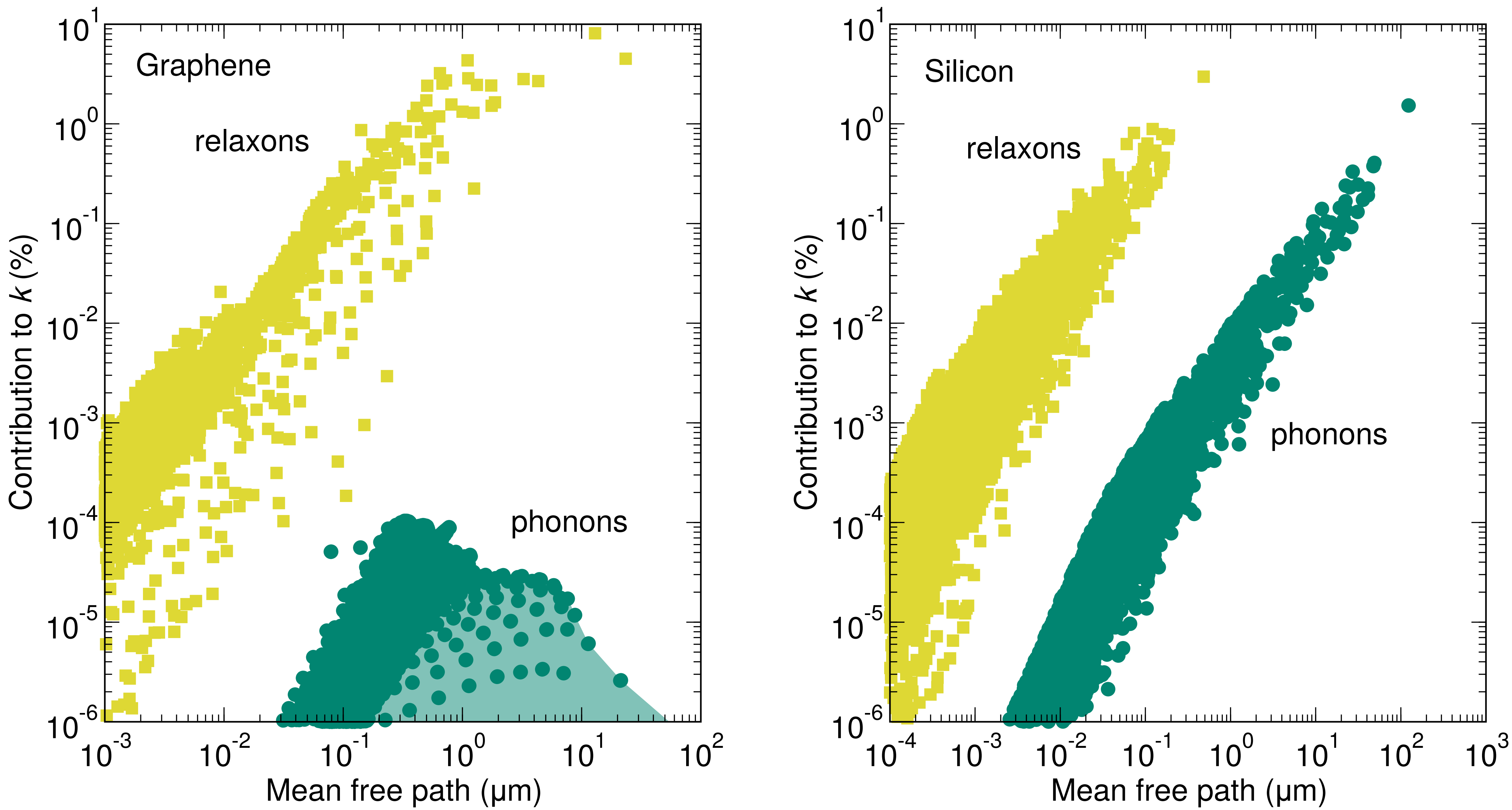
The theory changes the relevant time scale in graphene by orders of magnitude; and few modes (~20) contribute for most of transport

Velocities



The velocity of heat transport is not the speed of sound (20km/s in graphene, 18km/s in silicon), but much smaller (0.1 - 1 km/s)

Mean free paths



The distances at which heat is dissipated by each mode is very different from the phonon mean free paths.

Surface scattering

[*Nano Letters ASAP, (2017)*]

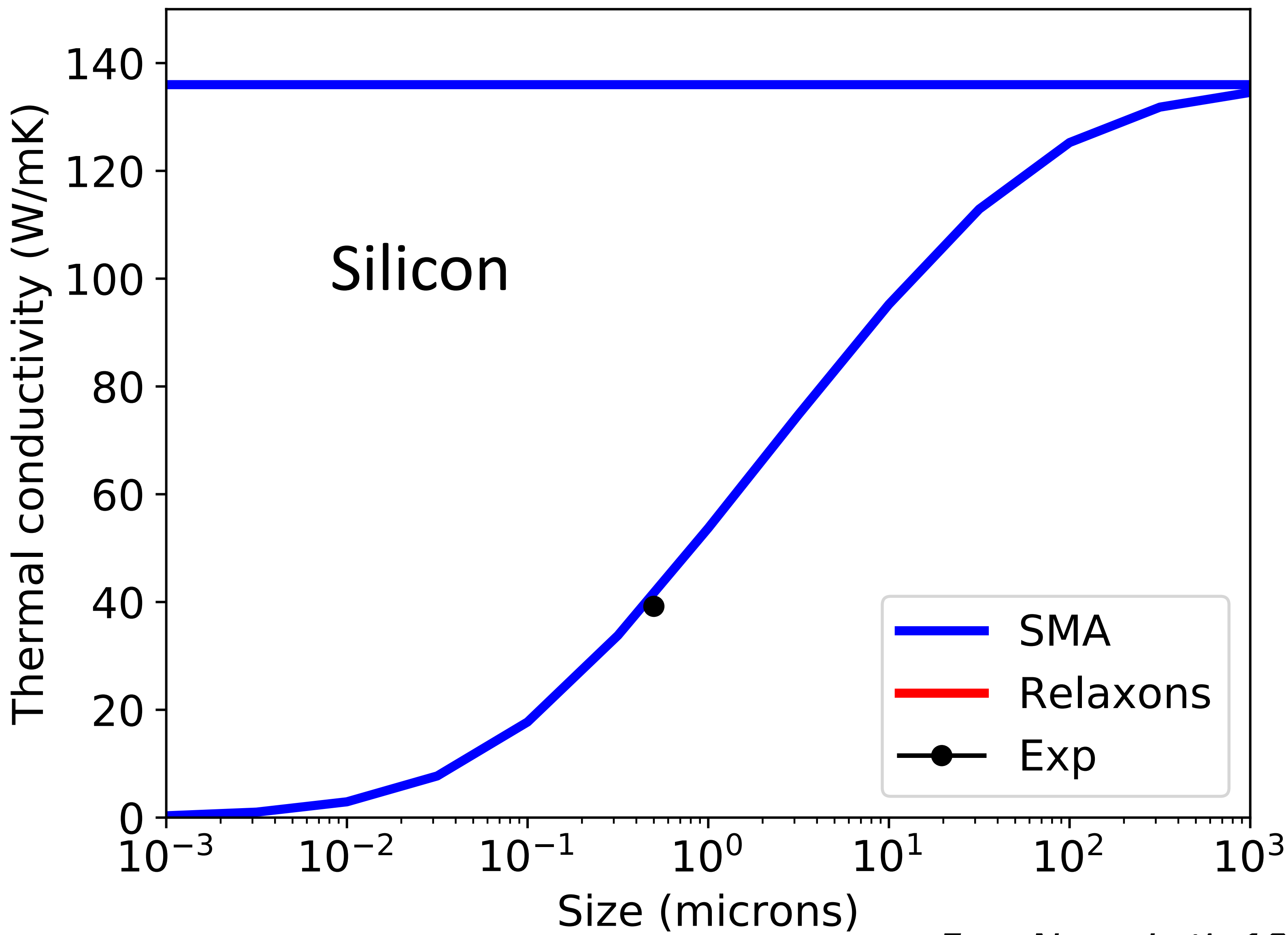
Typical interpretation: a phonon only travels for (1) the distance between scattering events or (2) the sample length *L*.
Therefore, the effective phonon relaxation time is:

$$\frac{1}{\tau_\mu} \to \frac{1}{\tau_\mu} + \frac{v_\mu}{L} \qquad \Longleftrightarrow \qquad \frac{1}{\Lambda_\mu} \to \frac{1}{\Lambda_\mu} + \frac{1}{L}$$

Can we say the same for relaxons?

$$? \quad \begin{matrix} \dfrac{1}{\tau_\alpha} \to \dfrac{1}{\tau_\alpha} + \dfrac{V_\alpha}{L} \\[2ex] \dfrac{1}{\Lambda_\alpha} \to \dfrac{1}{\Lambda_\alpha} + \dfrac{1}{L} \end{matrix} \quad ?$$

Let's see…



Exp: Nano Lett. 107, 11 (2011)

In silicon, the traditional approach works for phonons, but results for relaxons are off by two orders of magnitude.

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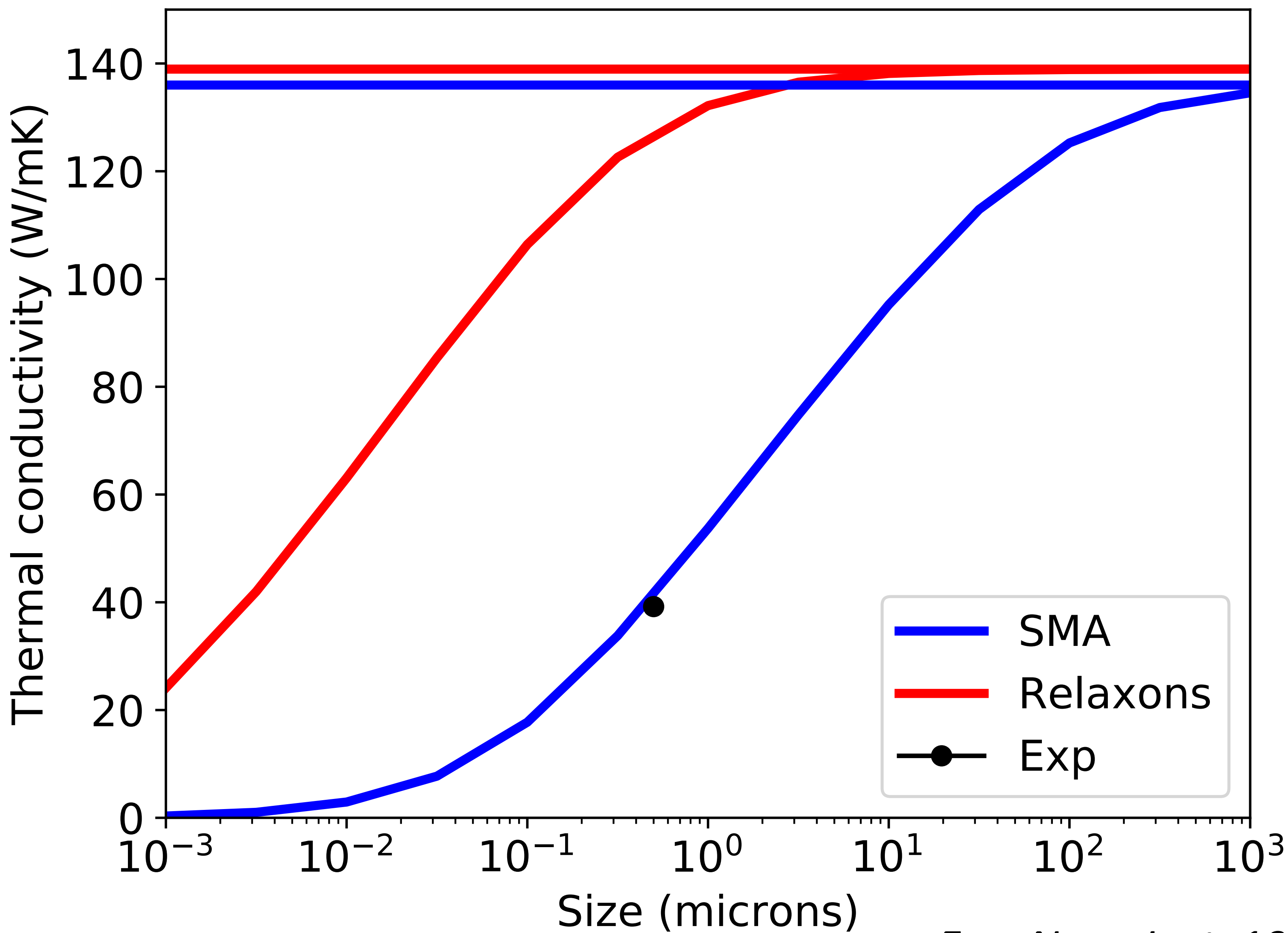

Exp: Nano Lett. 107, 11 (2011)

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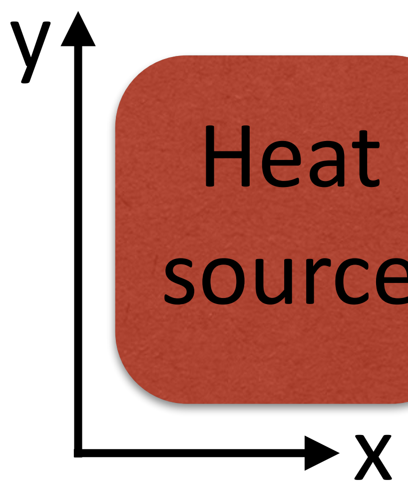
Question: why do we use this relation in first place? [*Phys. Rev. 33, 92 (1961)*]

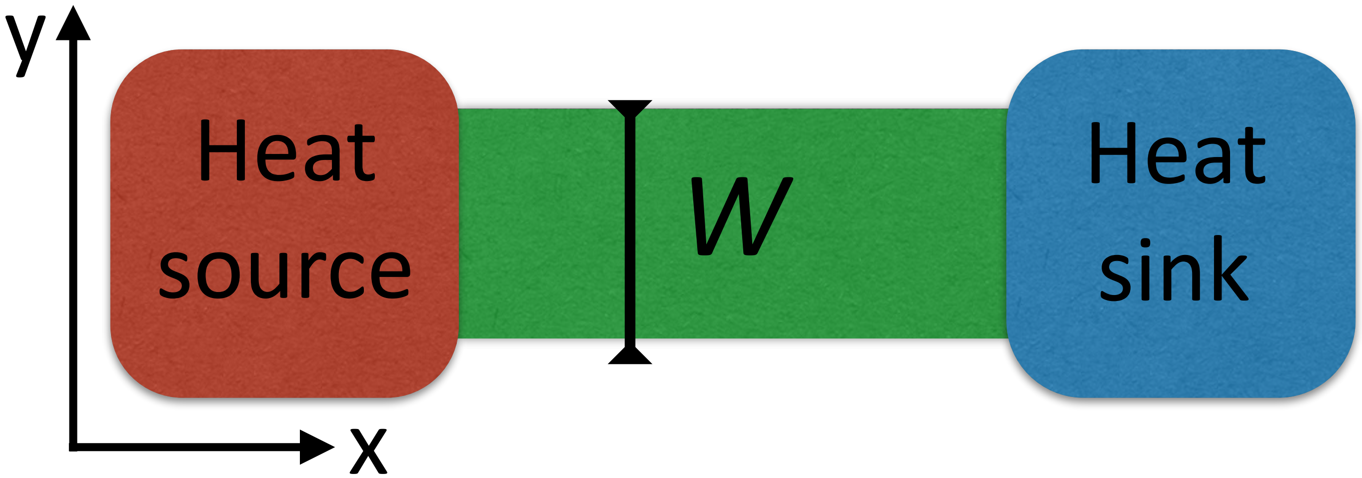
$$\frac{1}{\tau_\mu} \rightarrow \frac{1}{\tau_\mu} + \frac{v_\mu}{L}$$

SMA surface scattering



We study a 2D ribbon, of finite width but infinite length

SMA surface scattering



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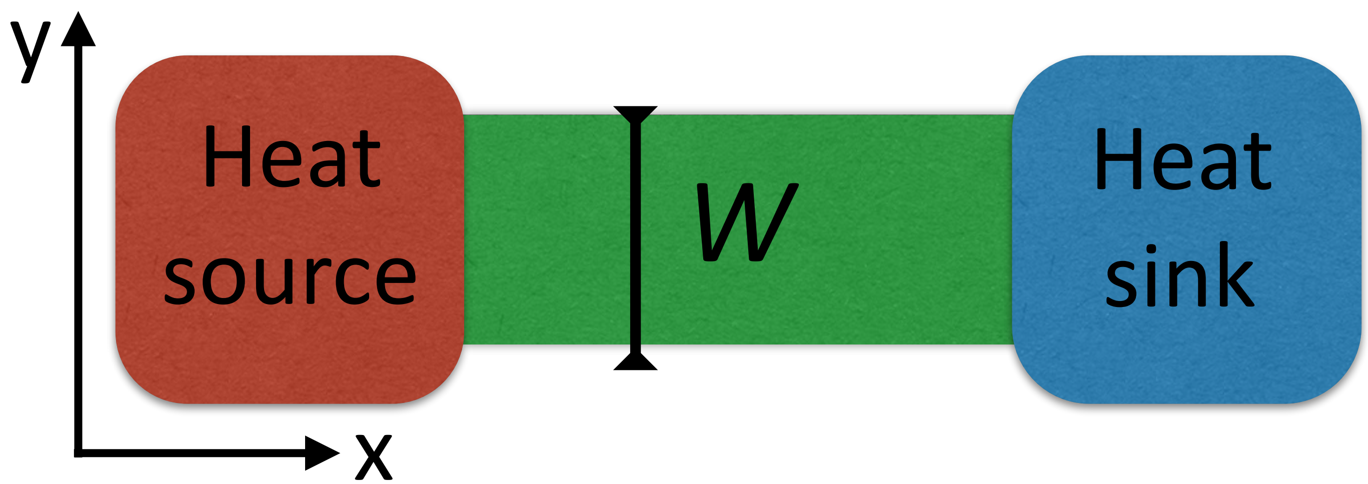
$$n_\mu = \bar{n}_\mu + \Delta n_\mu$$

SMA

$$\cancel{\frac{\partial n_\mu}{\partial t}} + \boldsymbol{v}_\mu \cdot \boldsymbol{\nabla} n_\mu = -\frac{1}{\mathcal{V}} \sum_{\mu'} \Omega_{\mu\mu'} \Delta n_{\mu'}$$

SMA surface scattering



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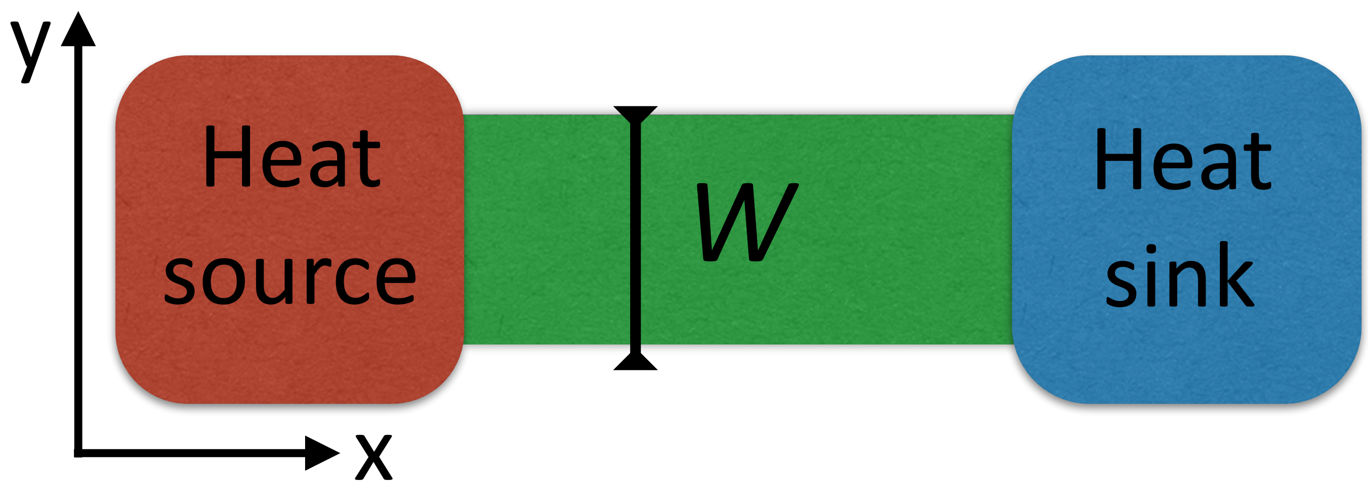
$$\boldsymbol{v}_\mu \cdot \boldsymbol{\nabla} T \frac{\partial \bar{n}_\mu}{\partial T} + \boldsymbol{v}_\mu \cdot \boldsymbol{\nabla} \Delta n_\mu = -\frac{\Delta n_\mu}{\tau_\mu}$$

SMA surface scattering



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$$\boldsymbol{v}_\mu \cdot \boldsymbol{\nabla} T \frac{\partial \bar{n}_\mu}{\partial T} + \boldsymbol{v}_\mu \cdot \boldsymbol{\nabla} \Delta n_\mu = -\frac{\Delta n_\mu}{\tau_\mu}$$

A finite sizes system doesn't have translational invariance: $\Delta n_\mu(\boldsymbol{r})$ depends on space. In the chosen geometry, we have:

$$v_\mu^y \frac{\partial \Delta n_\mu(y)}{\partial y} + \frac{\partial \bar{n}_\mu}{\partial T} v_\mu^x = -\frac{\Delta n_\mu(y)}{\tau_\mu}$$

SMA surface scattering

We must solve the following BTE:

$$\lambda_\mu^y \frac{\partial \Delta n_\mu(y)}{\partial y} + \Delta n_\mu(y) = -\frac{\partial \bar{n}_\mu}{\partial T} \lambda_\mu^x$$

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Case 1: $v^y{}_\mu = 0$: the phonon travels parallel to the surface;
it's population is the same as in the bulk case;

$$\Delta n_\mu(y) = \Delta n_\mu^{\text{bulk}}$$

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Case 2: $v^y{}_\mu > 0$ is more involved (the case $v^x{}_\mu < 0$ is similar).

$$\Delta n_\mu(y) = \Delta n_\mu^{\text{bulk}} + c_\mu e^{-y/\lambda_\mu^y}$$

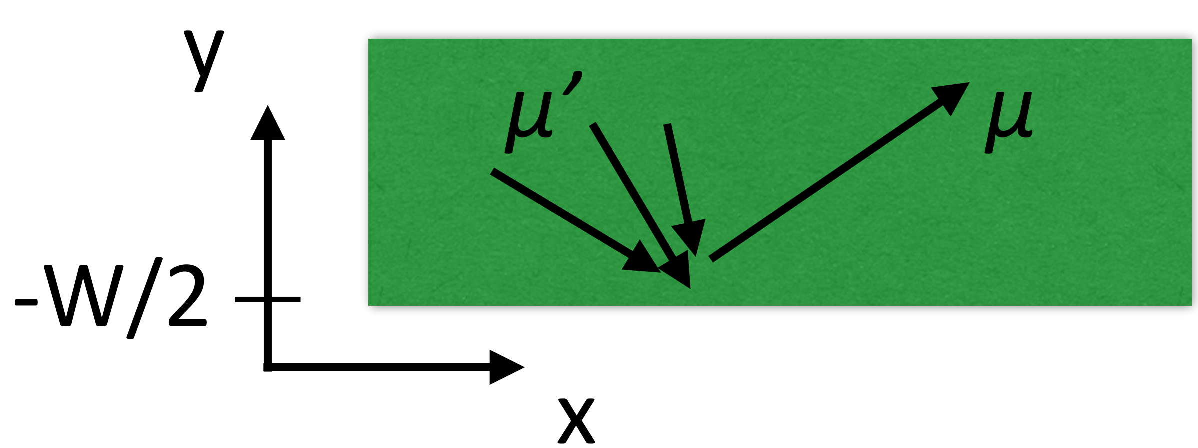
SMA surface scattering

$$\Delta n_\mu = \Delta n_\mu^{\text{bulk}} + c_\mu e^{-y/\lambda_\mu^y}$$

We must use a boundary condition. Ziman proposed to relate it with the surface phonon reflectivity:

The number of phonons traveling away from the surface must be equal to the reflected number of phonons that were traveling against it:

$$\Delta n_\mu \left(y = -\frac{W}{2} \middle| v_\mu^y > 0\right) = \sum_{\mu'} R_{\mu'}^{\mu} \Delta n_{\mu'} \left(y = -\frac{W}{2} \middle| v_{\mu'}^y < 0\right)$$



SMA surface scattering

Suppose perfect absorbance (*R=0*):

$$\Delta n_\mu(y) = \Delta n_\mu^{\text{bulk}}\left(1 - e^{-\frac{y+W/2}{\lambda_\mu^y}}\right) \qquad v_\mu^y > 0$$

$$\Delta n_\mu(y) = \Delta n_\mu^{\text{bulk}}\left(1 - e^{-\frac{y-W/2}{\lambda_\mu^y}}\right) \qquad v_\mu^y < 0$$

$$\Delta n_\mu(y) = \Delta n_\mu^{\text{bulk}} \qquad v_\mu^y = 0$$

Finally, the thermal conductivity is:

$$k(y) = -\frac{Q(y)}{\nabla T} = -\frac{1}{V}\sum_\mu v_\mu^x \hbar\omega_\mu \Delta n_\mu(y) = k^{\text{bulk}} - \Delta k^{\text{surf}}(y)$$

Note: thermal conductivity is not a bulk property!

SMA surface scattering

Final step: if we want to neglect the detailed space-dependence, we can average results in space:

$$\frac{\partial \bar{n}_\mu}{\partial T} v_\mu^x = -(\frac{1}{\tau_\mu^b} + \frac{1}{\tau_\mu})\Delta n_\mu$$

$$\Delta n_\mu = \frac{1}{W}\int_{-W/2}^{W/2} \Delta n_\mu(y) dy$$

$$\frac{1}{\tau_\mu^b} = \frac{\int_{-W/2}^{W/2} v_\mu^y \frac{\partial \Delta n_\mu(y)}{\partial y} dy}{\int_{-W/2}^{W/2} \Delta n_\mu(y) dy} \approx \frac{v_\mu^y}{L}$$

Take home message: surface effects must be studied in real space. Results can be written in reciprocal space, but after averaging over space

Surface scattering

Let's do the same with relaxons. We start from the BTE:

$$\sqrt{\frac{C}{k_B T^2}} V_{0\alpha}^{(x)} + \sum_{\beta} V_{\alpha\beta}^{(y)} \frac{\partial \tilde{f}_\beta}{\partial y} = -\frac{\tilde{f}_\alpha}{\tau_\alpha}$$

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We make another auxiliary scaling ($g_\alpha = \frac{1}{\sqrt{\tau_\alpha}} \tilde{f}_\alpha$) and get:

$$\sum_\beta \Lambda_{\alpha\beta}^{(y)} \frac{\partial g_\beta}{\partial y} = -g_\alpha + g_\alpha^\infty$$

g_α^∞ : bulk solution

$$V_{\alpha\beta}^{(y)} = \frac{1}{V} \sum_\mu \theta_\mu^\alpha v_\mu^y \theta_\mu^\beta$$

$\Lambda_{\alpha\beta}^{(y)} = \sqrt{\tau_\alpha} V_{\alpha\beta}^{(y)} \sqrt{\tau_\beta}$: matrix with dimensionality of length

It's a differential equation that can be solved analytically!

Surface scattering

The equation is trivial to solve in the basis of the eigenvectors of the matrix Λ:

$$\sum_\beta \Lambda^{(y)}_{\alpha\beta}\psi_{\beta i} = \lambda^{(y)}_i \psi_{\alpha i}$$

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$$g_\alpha = \sum_i \psi_{\alpha i} d_i$$

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Find a simpler differential equation

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With solutions

$$d_i = d_i^\infty + c_i e^{-y/\lambda^{(y)}_i}$$

Same as before, but in a different basis

Surface scattering

Let's think at the modes $\psi_{\alpha i}$ as particles moving towards $y>0$ (if $\lambda_i^y>0$) or $y<0$ (if $\lambda_i^y<0$). The boundary condition is fixed like before:

$$\delta_i = d_i\left(y = -\frac{W}{2}\right)\Big|_{\lambda_i^{(y)}>0} = \sum_j \mathcal{R}_i^j d_j\left(y = -\frac{W}{2}\right)\Big|_{\lambda_j^{(y)}<0}$$

If there's no reflection at the surface ($R=0$), we find an analytical solution of the BTE:

$$d_i = d_i^\infty - d_i^\infty e^{-(y+\frac{W}{2})/\lambda_i^{(y)}} \qquad \lambda_i^y > 0$$

$$d_i = d_i^\infty - d_i^\infty e^{-(y-\frac{W}{2})/\lambda_i^{(y)}} \qquad \lambda_i^y < 0$$

$$d_i = d_i^\infty \qquad \lambda_i^y = 0$$

Surface scattering

With a little more algebra, finally, the thermal conductivity is:

$$k(y) = -\frac{Q}{\nabla T}$$

$$= \sum_{\alpha} C V_{\alpha}^{(x)} \Lambda_{\alpha}^{(x)} - k_B T^2 \sum_{\alpha\beta} g_{\alpha}^{\infty} \Bigg(\sum_{\lambda_i^{(y)}>0} \psi_{\alpha i} e^{-\frac{y+\frac{W}{2}}{\lambda_i^{(y)}}} \psi_{i\beta}^{T} + \sum_{\lambda_i^{(y)}<0} \psi_{\alpha i} e^{-\frac{y-\frac{W}{2}}{\lambda_i^{(y)}}} \psi_{i\beta}^{T} \Bigg) g_{\beta}^{\infty}$$

$$= k^{\infty} - \Delta k^{\mathrm{surf}}(y)$$

- The conductivity is a bulk term minus a surface term;
- Conductivity isn't a bulk property: depends on where it is measured, and, in general, on the shape of the material;
- *The length scale of surface scattering is determined by the eigenvalues of $\Lambda_{\alpha\beta}$, not by the relaxon mean free paths.*

$$\frac{1}{\tau_\alpha} \rightarrow \frac{1}{\tau_\alpha} + \frac{V_\alpha}{L}$$

Surface scattering

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$$
\begin{aligned}
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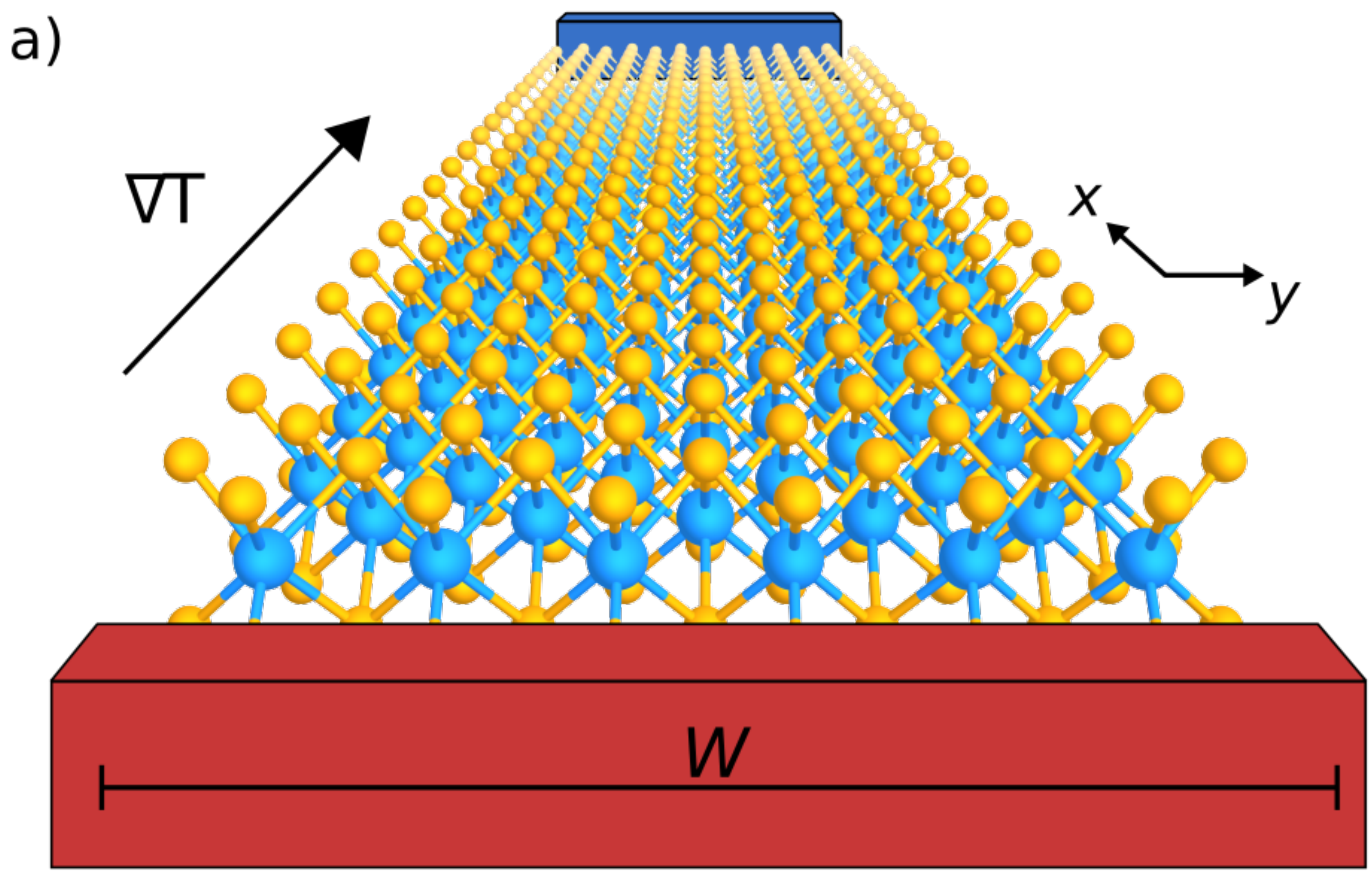
MoS_2 monolayer

Ribbon geometry (finite width, infinite length)



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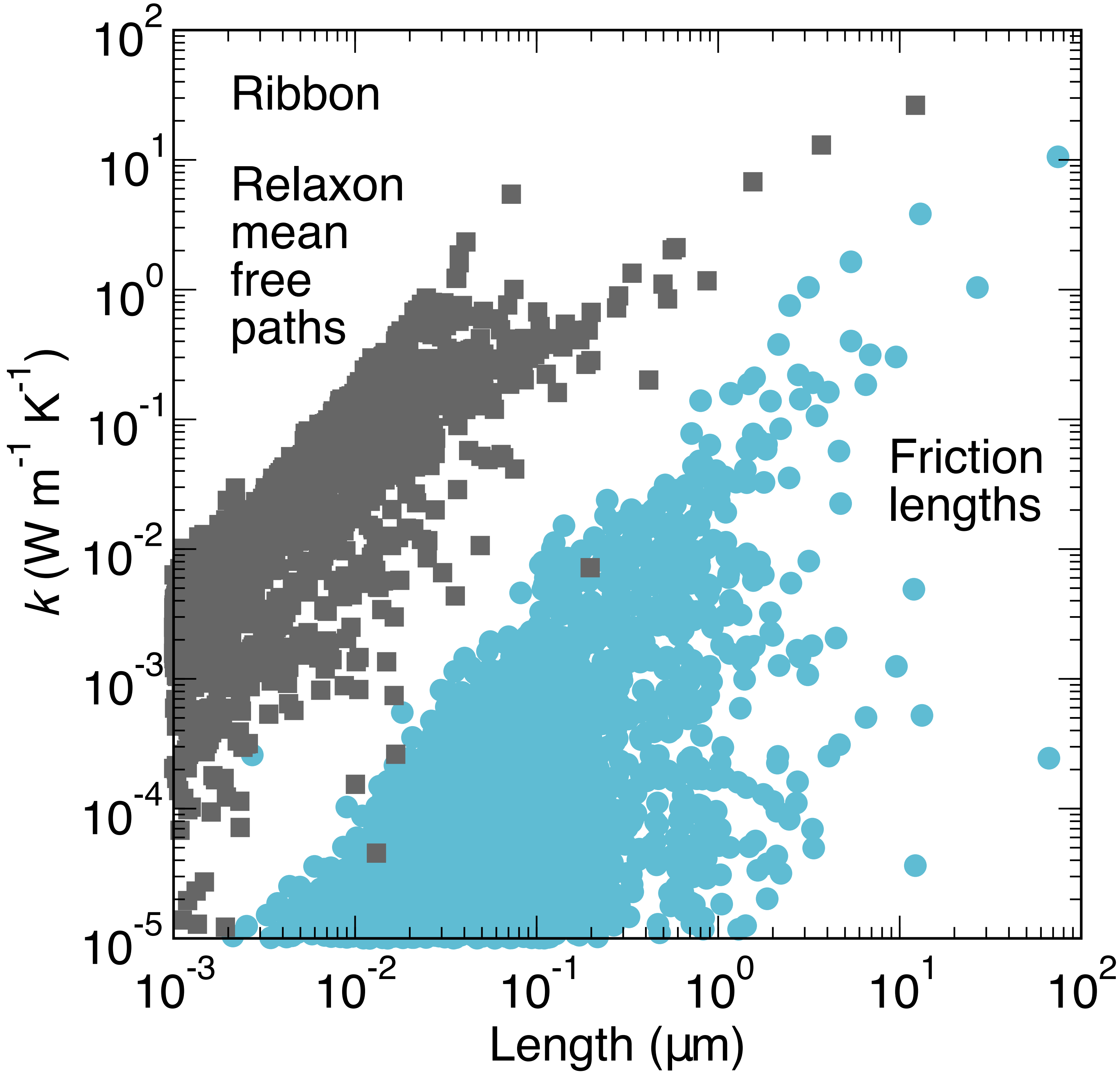
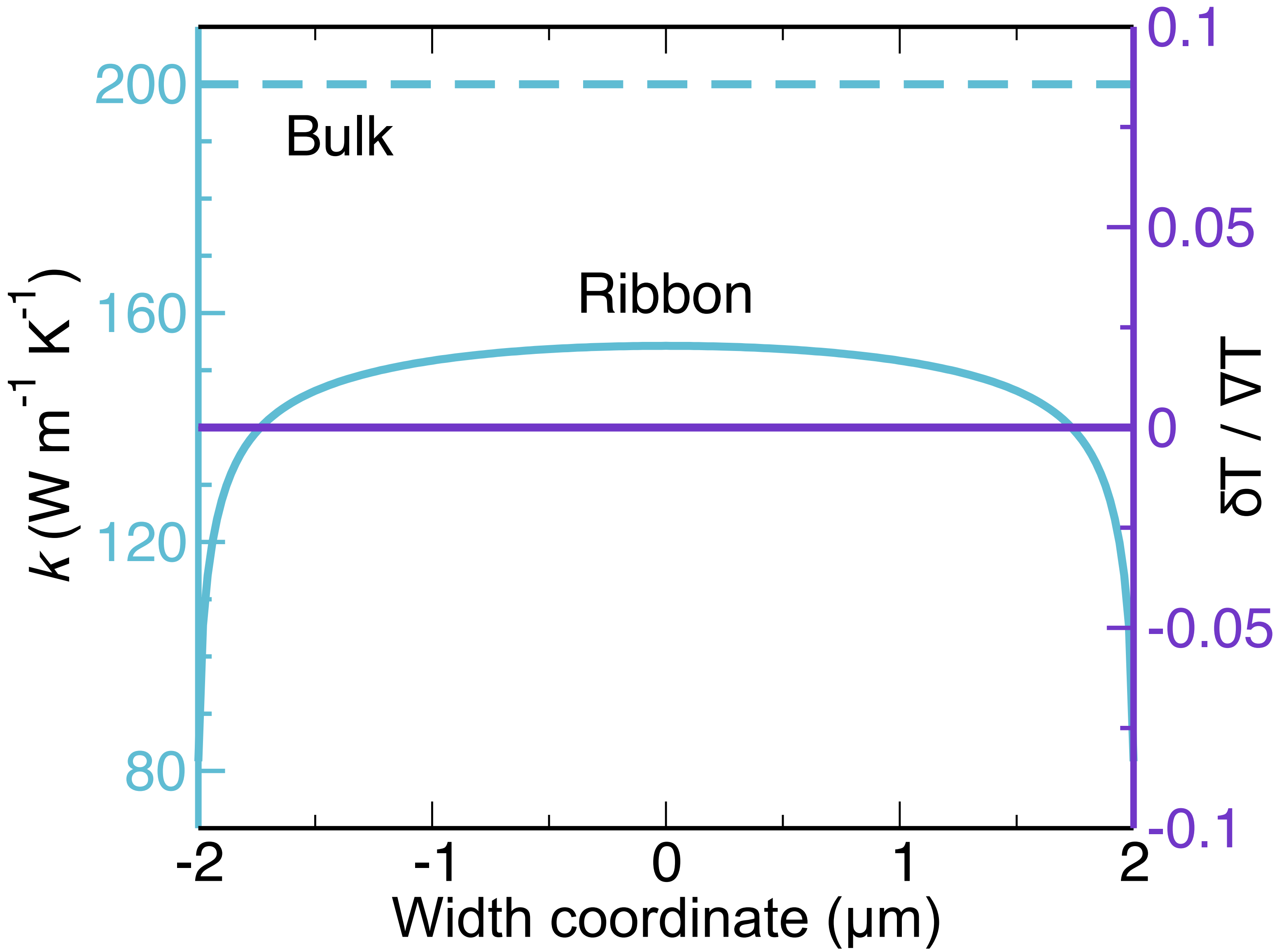
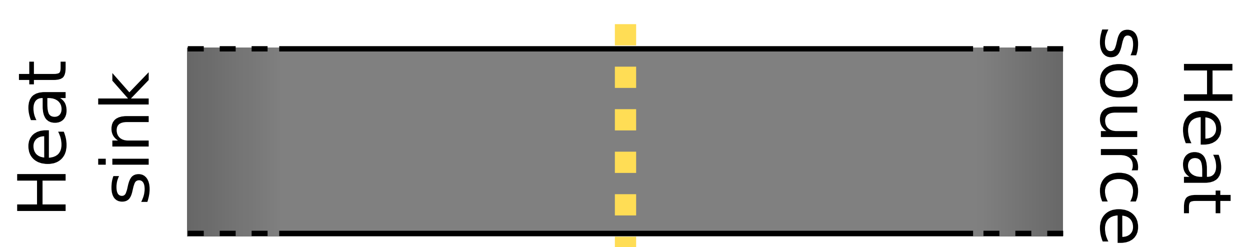


Contribution to thermal conductivity from relaxon $\vartheta_{\mu\alpha}$ and reduction of thermal condutivity from mode $\psi_{\alpha i}$

Friction lengths are much longer than carrier mean free paths.

MoS_2 monolayer





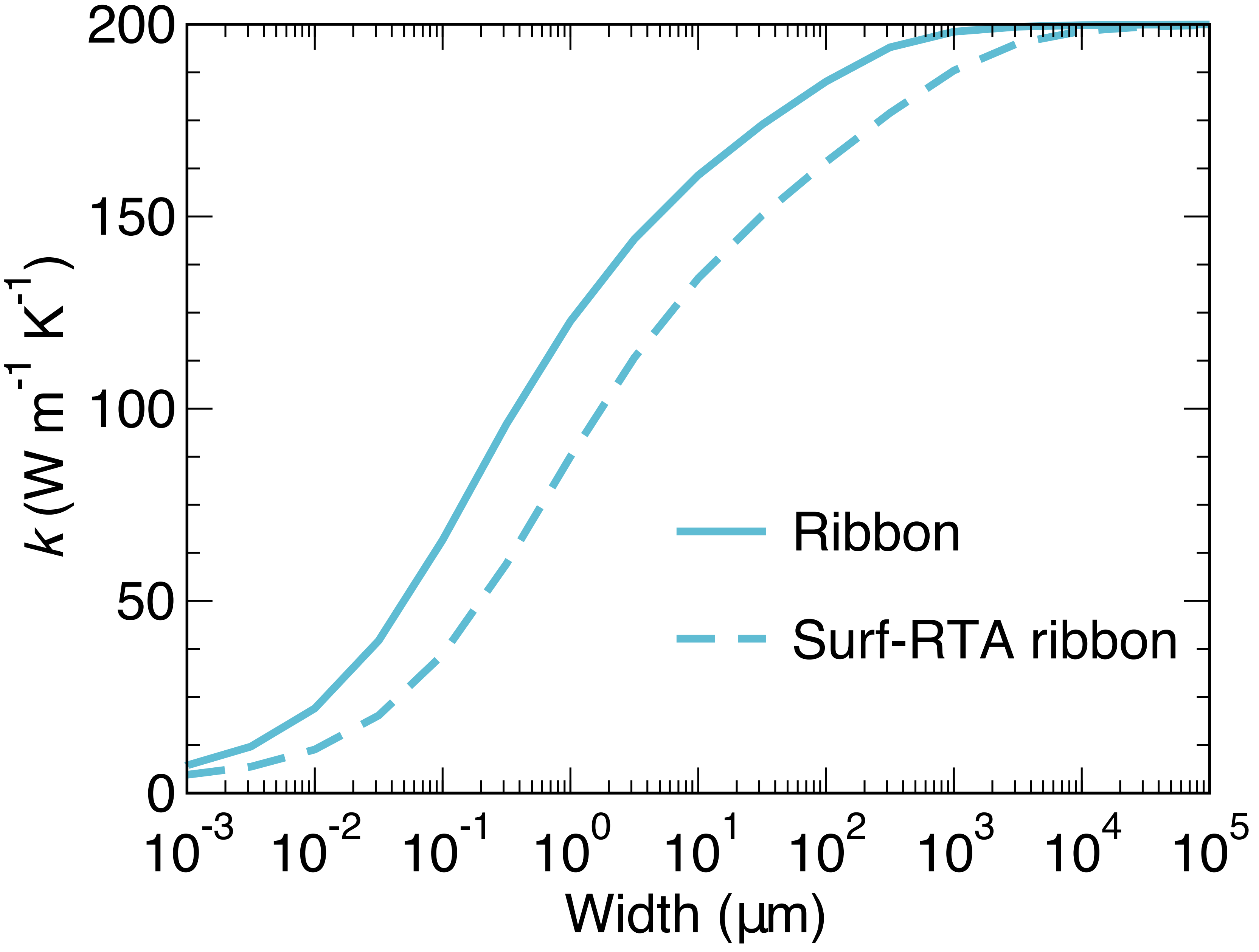
Thermal conductivity has a minimum at the surfaces and maximum at the center.

Temperature response ($\Delta T = \Delta E/C$) is constant along the ribbon width.

The same behavior is seen in liquids (e.g. rivers): phonons display hydrodynamic behaviors

Hence, identify the eigenvalues λ_i^y as "friction lengths"

MoS_2 monolayer

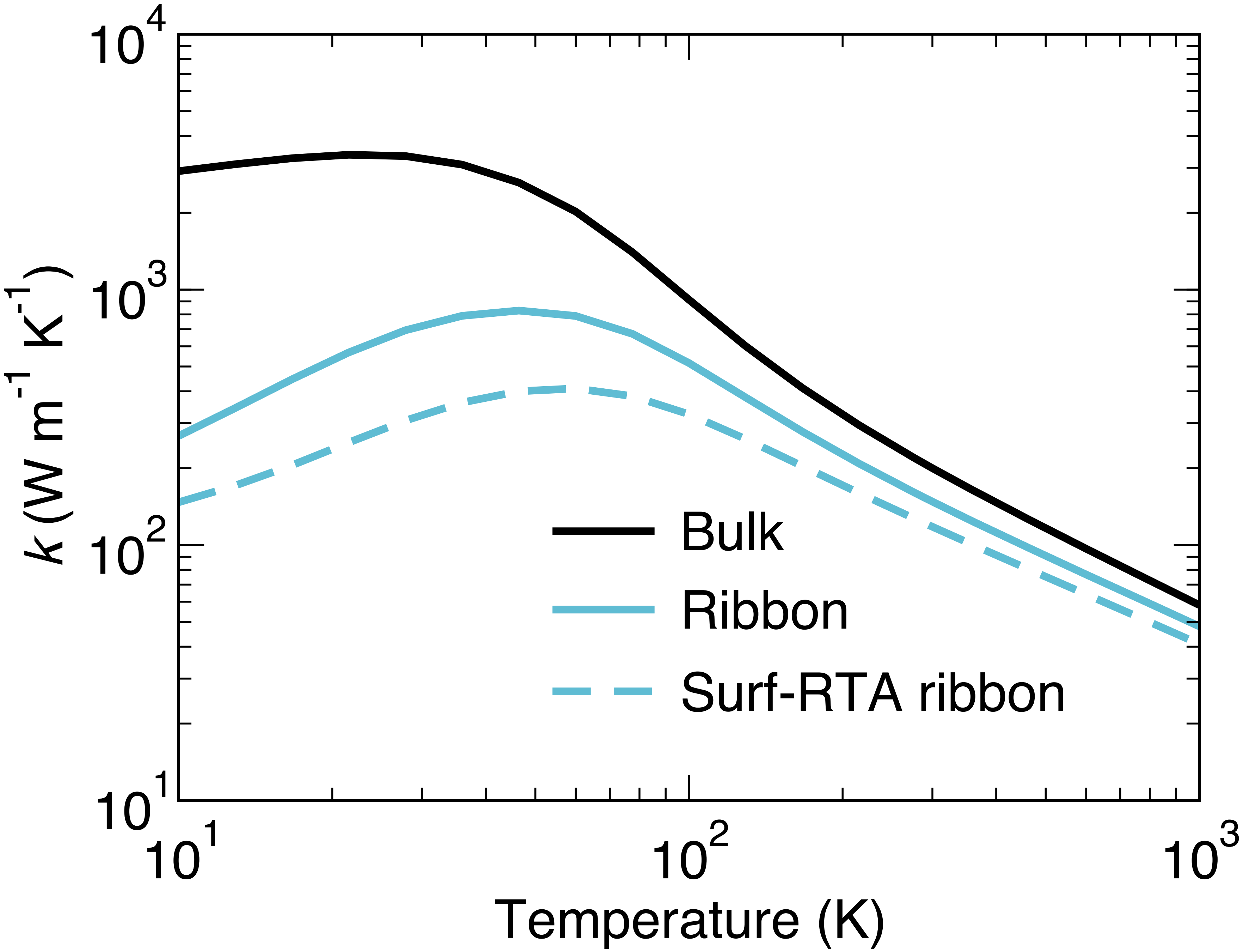


Surf-RTA is a model of surface scattering often seen in literature

$$\Omega_{\mu\mu'} \rightarrow \Omega_{\mu\mu'} + \delta_{\mu\mu'} \frac{v_\mu}{L}$$

This mixes SMA and non-SMA results and it's incorrect
(1) theoretically and
(2) by an order of magnitude (the horizontal translation)

MoS_2 monolayer



Trends follow the expected behavior;

The difference is bigger at low temperatures, where surface scattering is more important;

Note: the log-scale compresses differences!

Second sound

[*arXiv:1612.04317*]

The Boltzmann transport equation admits wave-like solutions:

$$\Delta n_\mu = \sum_{\boldsymbol{k}\alpha} c_\alpha I_\mu^\alpha e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega_\alpha t)}$$

The Boltzmann equation is a non-hermitian eigenvalue problem

$$\frac{1}{V}\sum_{\mu'} \Omega_{\mu\mu'} I_{\mu'} + i\boldsymbol{k}\cdot\boldsymbol{v}_\mu I_\mu = i\omega I_\mu$$

Waves have a dispersion relation, with complex frequencies:

$$\omega = \omega_\alpha(\boldsymbol{k}) = \bar{\omega}_\alpha(\boldsymbol{k}) - \frac{i}{\tau_\alpha(\boldsymbol{k})}$$

$$\Delta n_\mu^\alpha(\boldsymbol{k}) = |I_\mu^\alpha(\boldsymbol{k})| e^{-t/\tau_\alpha(\boldsymbol{k})} \sin(\boldsymbol{k}\cdot\boldsymbol{r} - \overline{\omega}_\alpha(\boldsymbol{k})t + \phi)$$

Second sound

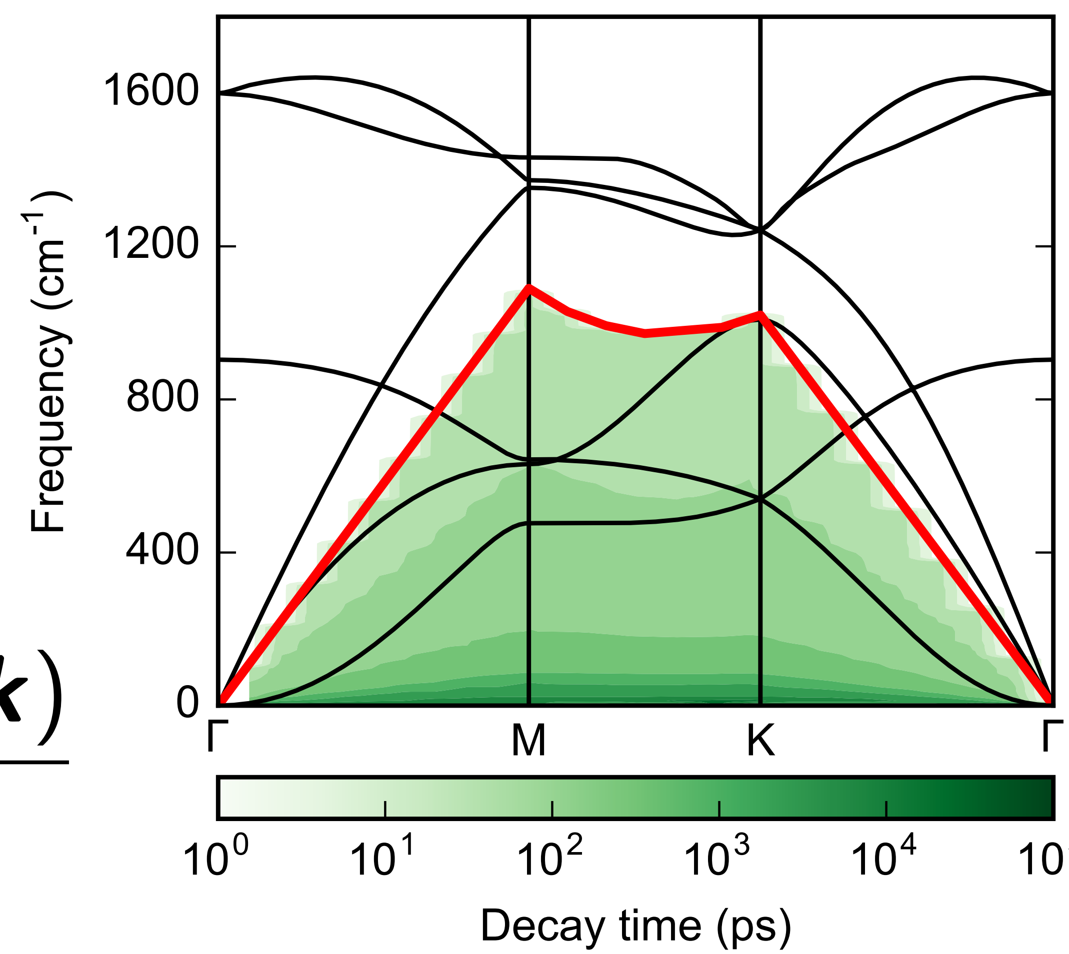
Population waves $\Longleftrightarrow$ Temperature waves

$$\Delta n_\mu^\alpha = I_\mu^\alpha e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega_\alpha(\boldsymbol{k})t)} \qquad \Delta E = \frac{1}{\mathcal{V}}\sum_\mu \hbar\omega_\mu \Delta n_\mu = C\Delta T$$

Using this Ansatz, we obtain damped wave solutions of temperature, i.e. a basis set of second sound modes

$$\frac{\partial^2 T}{\partial t^2} + \frac{1}{\tau_{ss}}\frac{\partial T}{\partial t} - (v_{ss})^2\nabla^2 T = 0$$

$$\omega_\alpha(\boldsymbol{k}) = \bar{\omega}_\alpha(\boldsymbol{k}) - \frac{i}{\tau_\alpha(\boldsymbol{k})} \qquad v_{ss} = \frac{\partial\bar{\omega}_\alpha(\boldsymbol{k})}{\partial\boldsymbol{k}}$$



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- A kinetic theory of the relaxon gas describes exactly thermal transport in crystals, revising time and length scales of transport.
- Surface "scattering" is in fact an effect of the friction (viscosity) of the 'liquid' of vibrations.