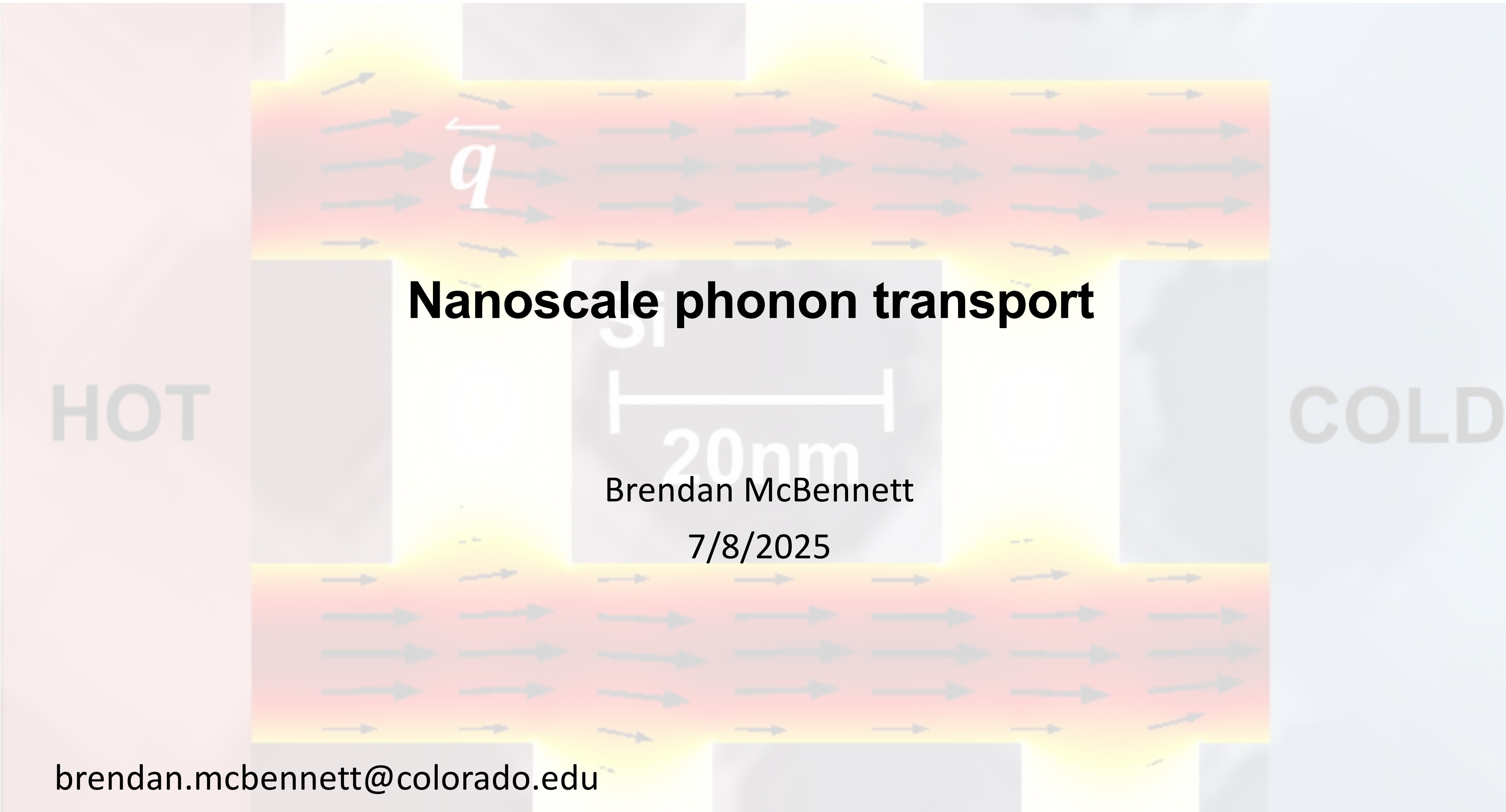


# Nanoscale phonon transport

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7/8/2025

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# Acknowledgements


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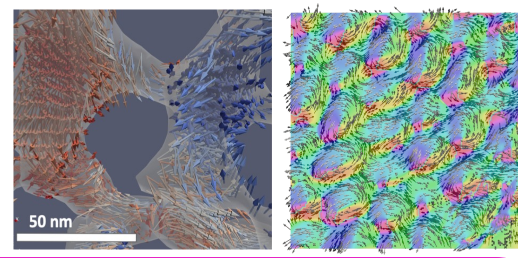
Javier Bafaluy
Lluc Sendra
Weinan Chen
Ismaila Dabo
Mahmoud Hussein

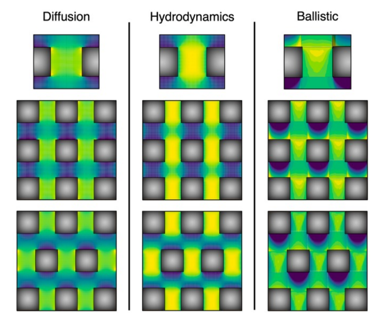
UAB
Universitat Autònoma de Barcelona

# Outline

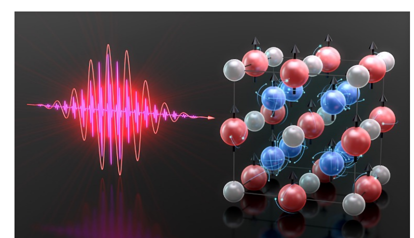
- **Lecture 1 – Brief intro to the physics of High Harmonics (Nobel 2023) and coherent imaging**




- **Lecture 2 – Nanoscale phonon transport** (with Dr. Brendan McBennett, NIST)




- **Lecture 3 – Understanding spin and many-body electron-phonon dynamics** via EUV MOKE and ARPES



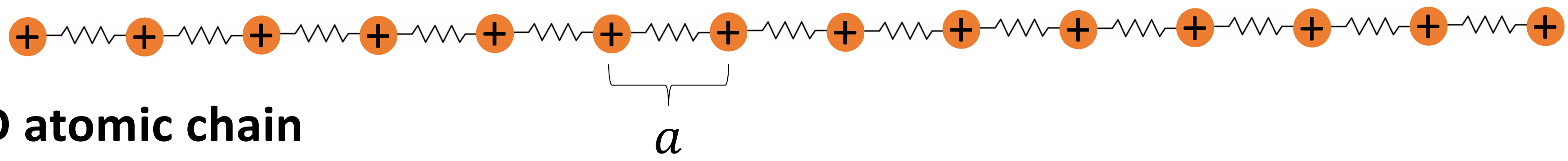
# Today’s outline

**1. Phonons overview**

2. Kinetic transport theory

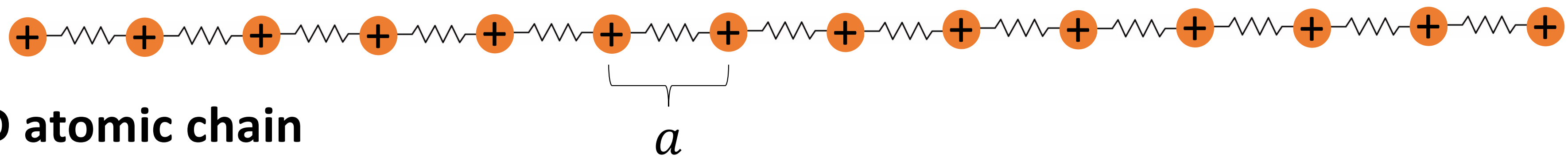
3. Phonon transport at the nanoscale

# Phonons




**1D atomic chain**

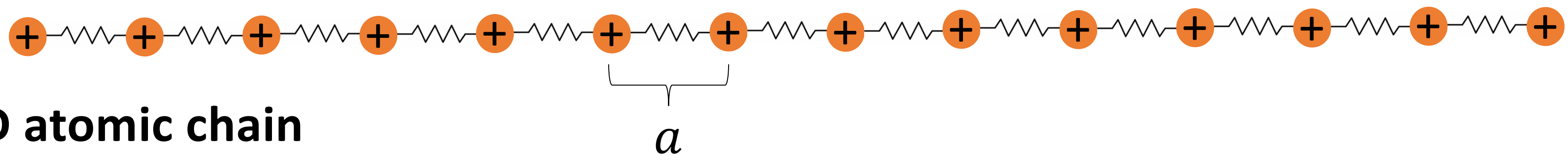
# Phonons




**1D atomic chain**

- $N$ atoms of mass $m$
- Lattice constant $a = 1$
- $u_n$ is displacement of atom $n$
- Periodic boundary conditions $u_n = u_{n+N}$

# Phonons

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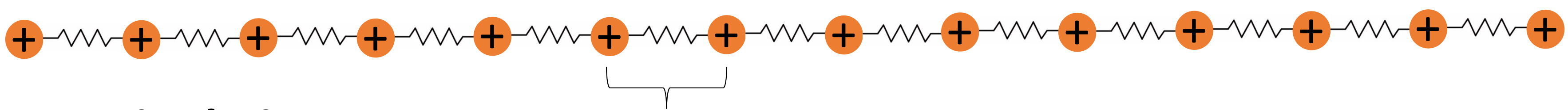
$$H = \frac{m}{2}\sum_n \dot{u}_n^2 + \omega_0^2 (u_n - u_{n-1})^2$$

$$\ddot{u}_n = \omega_0^2 \left(u_{n+1} + u_{n-1} - 2u_n\right)$$

# Phonons

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$$u_n \sim e^{ikx - i\omega_k t}$$

$$H = \frac{m}{2}\sum_n \dot{u}_n^2 + \omega_0^2 (u_n - u_{n-1})^2$$

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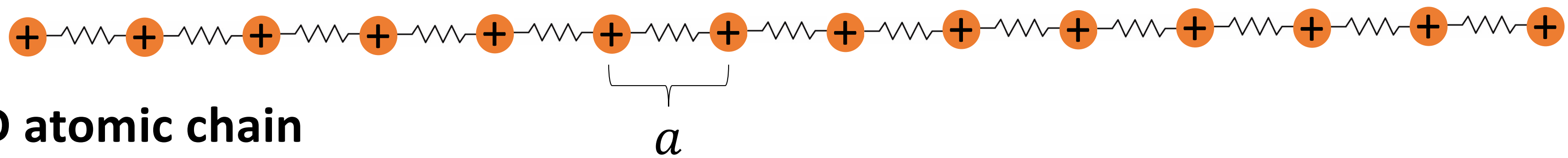
# Phonons

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$$u_n \sim e^{ikx - i\omega_k t}$$

$$\Rightarrow \quad k = \frac{2\pi}{N}(0, \pm 1, \pm 2 \ldots)$$

$$H = \frac{m}{2}\sum_n \dot{u}_n^2 + \omega_0^2 (u_n - u_{n-1})^2$$

$$\boxed{\omega_k = 2\omega_0 \sin\left|\frac{k}{2}\right|}$$

$$\ddot{u}_n = \omega_0^2 \,(u_{n+1} + u_{n-1} - 2u_n)$$

# Phonons

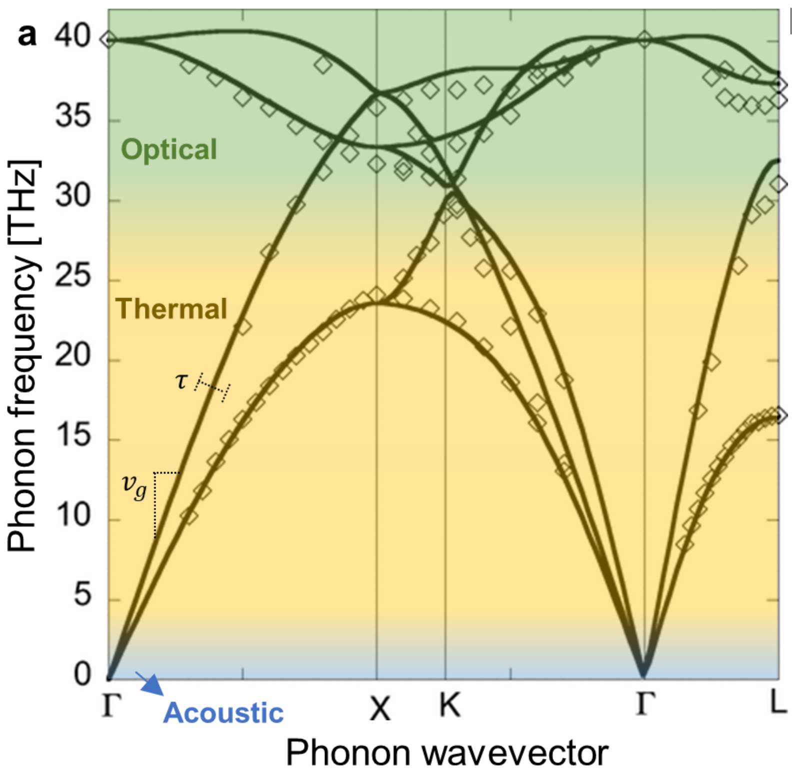
**Quantum mechanical approach**

$$H = \frac{m}{2}\sum_k \omega_k \left(n_k + \frac{1}{2}\right)$$

- Superposition of harmonic oscillators
- $n_k$ "phonons" in mode $k$
- For N atoms in 3D have 3N modes

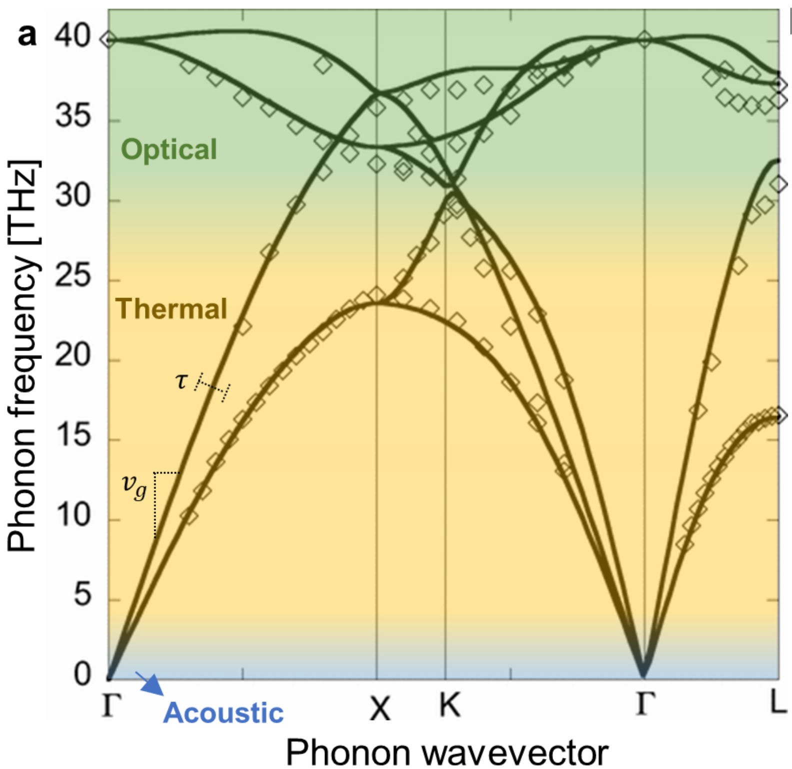
Phonons correspond to eigenstates of the **harmonic** Hamiltonian. Real crystals are not harmonic and so phonons will have finite lifetimes

# Phonons in real crystals




Phys. Rev. B **80**, 125203 (2009)

# Phonons in real crystals




**Acoustic phonons (sound)** – long wavelength, linear dispersion (kHz-GHz)

**Optical phonons** – very high frequency, low group velocity

**Thermal phonons** – heat carriers (THz)

**Mean free path:** $\Lambda = v\tau$

Phys. Rev. B **80**, 125203 (2009)

# Macroscopic thermal transport

$$\vec{q} = -\kappa \nabla T$$ Fourier's law

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T$$ Heat equation

| | |
|---|---|
| $\vec{q}$ | Heat flux |
| $\nabla T$ | Temperature gradient |
| $\kappa, \alpha$ | Thermal conductivity, diffusivity |

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How to relate macroscopic variables to microscopic quantities like $v_k, \tau_k, \omega_k, \Lambda_{\mathrm{k}}$?

# Today’s outline

1. Phonons overview

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# Distribution functions

- Treat phonons as particles (~1nm wavelengths at RT)
- Energy is conserved, but not much else
- $n_k(\boldsymbol{x}, t)$: number of phonons in mode $k$ at location $\boldsymbol{x}$ at time $t$
- In equilibrium, $n_k^0 = \left[e^{\hbar\omega_k/k_B T} - 1\right]^{-1}$ (phonons are bosons)

# Momentum conservation

- Phonons have zero average "real" momentum (the atoms in a crystal experience no net displacement as a phonon propagates)

- Instead they have crystal momentum $\hbar\vec{k}$

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- Crystal momentum is only conserved up to a reciprocal lattice vector $\vec{R}$
  - Normal collisions $\vec{k} + \vec{k}' = \vec{k}''$
  - Resistive (Umklapp) collisions $\vec{k} + \vec{k}' = \vec{k}'' + \vec{R}$

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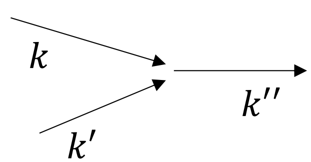
Image: Jacopo Bertolotti

# Boltzmann transport equation

$$\frac{\partial n_k(\boldsymbol{x},t)}{\partial t} + \underbrace{\vec{v}_k \cdot \nabla n_k(\boldsymbol{x},t)}_{\text{Drift}} = \underbrace{\mathrm{C}[n_k(\boldsymbol{x},t)]}_{\text{Collisions}}$$

$\mathrm{C}$ is the collision operator representing all the possible scattering events which can alter the distribution function. In general it is very difficult to calculate

$$\mathrm{C}[n_k(\boldsymbol{x},t)] \sim \int dk' dk'' |M(k,k',k'')|^2 \omega\omega'\omega'' \delta(\omega+\omega'-\omega'')(\mathrm{n}+1)(\mathrm{n}'+1)\mathrm{n}''$$



# Energy and heat flux

$$u(\boldsymbol{x}, t) = V^{-1} \sum_k \hbar\omega_k \, n_k(\boldsymbol{x}, t)$$

Energy density

$$\vec{q}(\boldsymbol{x}, t) = V^{-1} \sum_k \hbar\omega_k \vec{v}_k \, n_k(\boldsymbol{x}, t)$$

Heat flux density

These are very fundamental expressions – no assumptions yet

# Boltzmann to Fourier

$$\frac{\partial n_k(\boldsymbol{x},t)}{\partial t} + \vec{v}_k \cdot \nabla n_k(\boldsymbol{x},t) = \mathrm{C}[n_k(\boldsymbol{x},t)]$$

- Relaxation time approximation
- Local equilibrium approximation
- Steady state

$$\vec{q} = -\underbrace{\left(\sum_k v_k \Lambda_k c_k\right)}_{\text{thermal conductivity}} \nabla T$$

$\boxed{\text{Can calculate } v_k, \Lambda_k, c_k \text{ from first principles}}$

# Bulk thermal conductivity calculations

**Intrinsic lattice thermal conductivity of semiconductors from first principles**

D. A. Broido[a)]
*Department of Physics, Boston College, Chestnut Hill, Massachusetts 02467, USA*

M. Malorny and G. Birner
*Department of Physics, University of Regensburg, D-93040 Regensburg, Germany*

Natalio Mingo
*CEA-Grenoble, 17 Rue des Martyrs, Grenoble 38054, France*
*and Department of Electrical Engineering, University of California, Santa Cruz, California 95064, USA*

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*Cornell Nanoscale Facility, Cornell University, Ithaca, New York, 14853, USA*

(Received 30 October 2007; accepted 19 November 2007; published online 7 December 2007)

We present an *ab initio* theoretical approach to accurately describe phonon thermal transport in semiconductors and insulators free of adjustable parameters. This technique combines a Boltzmann formalism with density functional calculations of harmonic and anharmonic interatomic force constants. Without any fitting parameters, we obtain excellent agreement (<5% difference at room temperature) between the calculated and measured intrinsic lattice thermal conductivities of silicon and germanium. As such, this method may provide predictive theoretical guidance to experimental thermal transport studies of bulk and nanomaterials as well as facilitating the design of new materials. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2822891]

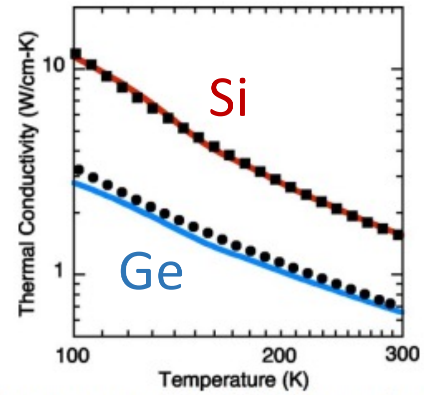


FIG. 1. (Color online) Lattice thermal conductivity $\kappa^{(i)}$ as a function of temperature. The red line and solid squares are the calculated and measured thermal conductivities of silicon, respectively, while the blue line and the solid circles are the corresponding quantities for germanium.

This works for bulk materials with dimensions ≫ mean free paths (100s of nm at room temperature). What about at the nanoscale?

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Length scales on the order of phonon mean free paths, time scales on the order of phonon lifetimes

# Example: second sound

$n_k = f(\omega_k, T)$

Arbitrary initial distribution function (ex. from ultrafast laser excitation)

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Arbitrary initial distribution function (ex. from ultrafast laser excitation)

Fast timescale

$$n_k = \left[e^{(\hbar\omega_k - \hbar\vec{k}\cdot\vec{u})/k_B T} - 1\right]^{-1}$$

Suitable intermediate distribution function

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Fast timescale

$$n_k = \left[e^{(\hbar\omega_k - \hbar\vec{k}\cdot u)/k_B T} - 1\right]^{-1}$$

Suitable intermediate distribution function

Slow timescale

$$n_k^0 = \left[e^{\hbar\omega_k/k_B T} - 1\right]^{-1}$$

Equilibrium distribution function

# Example: second sound

$$n_k = f(\omega_k, T)$$

Fast timescale

$$n_k = \left[e^{(\hbar\omega_k - \hbar\vec{k}\cdot u)/k_B T} - 1\right]^{-1}$$

$$\tau_{ss}\frac{\partial T}{\partial t} + \frac{\partial T}{\partial t} = \alpha\nabla^2 T$$

Damped wave equation for temperature!

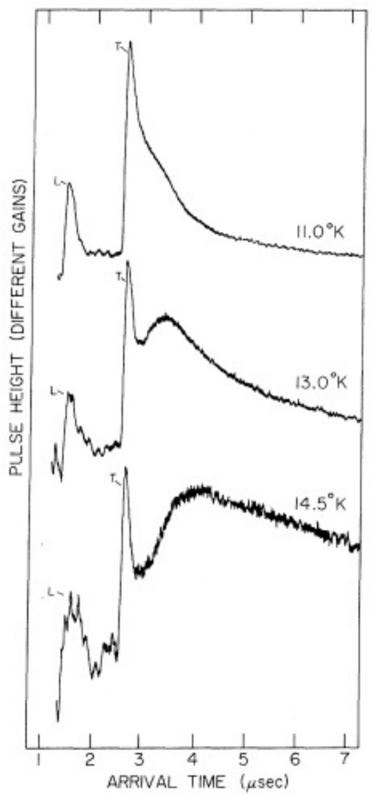
Slow timescale

$$n_k^0 = \left[e^{\hbar\omega_k/k_B T} - 1\right]^{-1}$$

$$\frac{\partial T}{\partial t} = \alpha\nabla^2 T$$ Heat equation

# Example: second sound

NaF, 13K (1971)




Phys. Rev. B **3** 1428 (1971)

# Example: second sound

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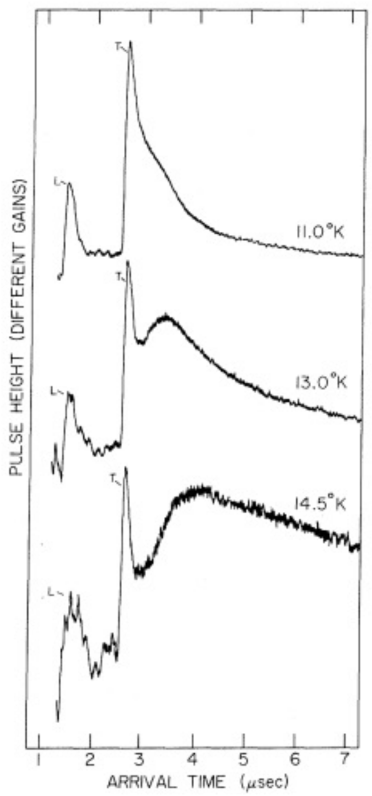
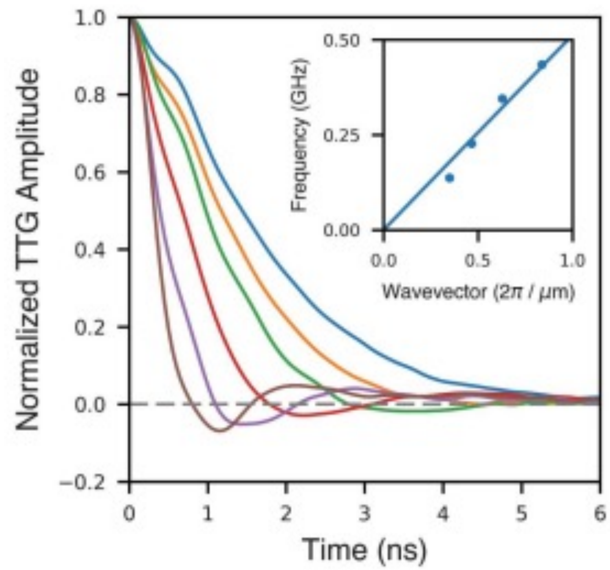
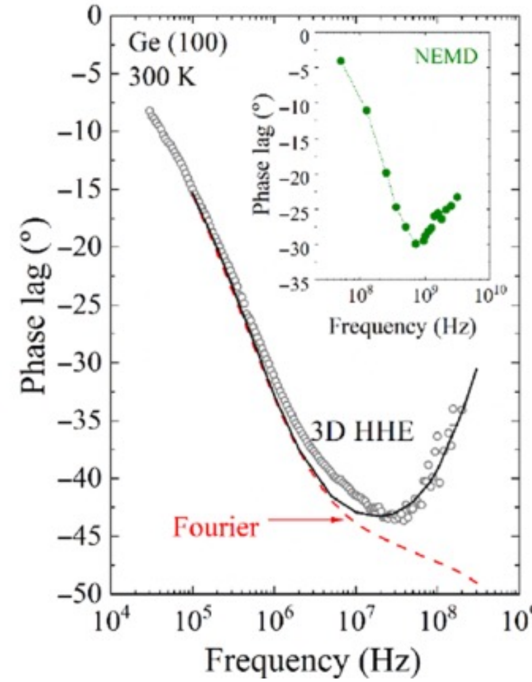



Graphite, 100K (2019)




1970s: exotic
2010s: 2D materials, higher temperatures

Ge, 300K (2021)




Phys. Rev. B **3** 1428 (1971)
Science 364, 375–379 (2019)
Sci. Adv **7** eabg4677 (2021)

# Example: confinement

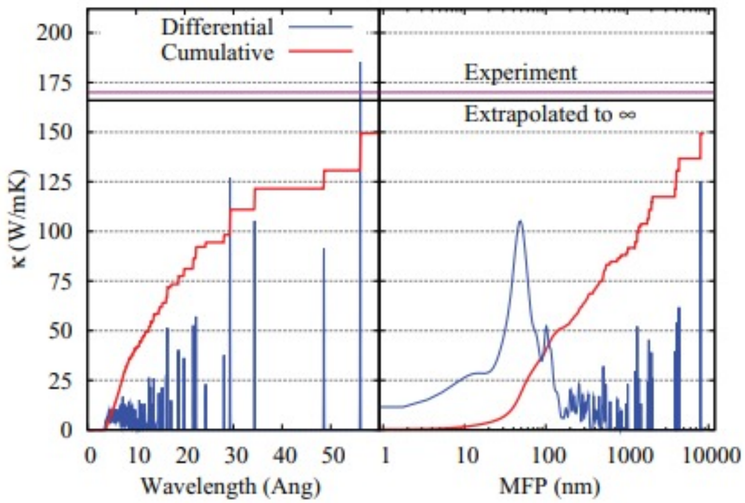



FIG. 6. (Color online) Cumulative contributions of phonons to the thermal conductivity at 277 K from the $18 \times 18 \times 18$ $k$-point mesh data. Left plot is according to the wavelengths, and right plot is according to the MFPs. Both differential and cumulative thermal conductivities are shown in blue and red, respectively. For comparison, the extrapolated (to infinite $k$-point mesh) and experimental $\kappa$ are also shown with horizontal lines at 166 and 174 W/mK, respectively.

- Phonon mean free paths in bulk silicon are 10s of nm to microns

Phys. Rev B **84**, 085204 (2011)

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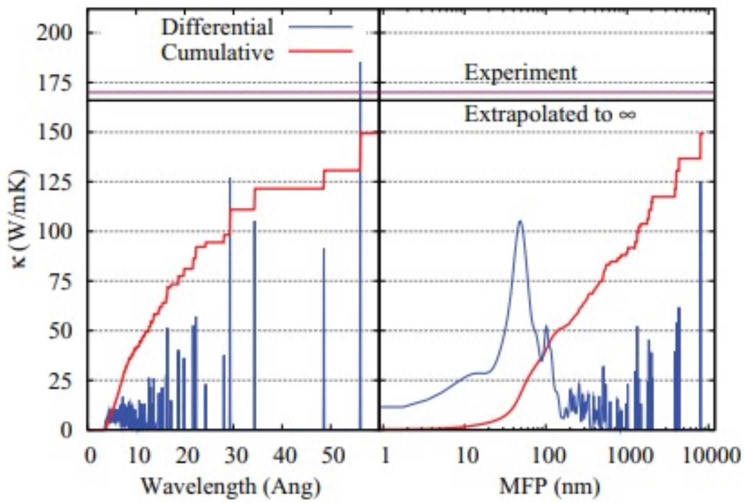


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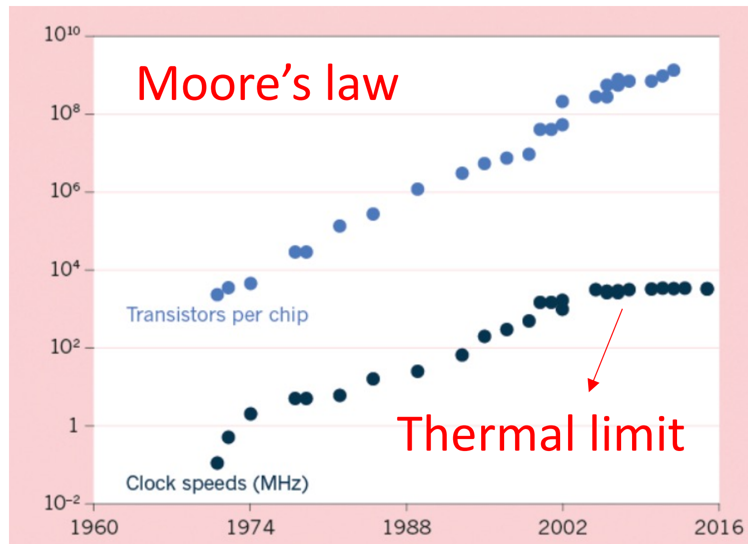
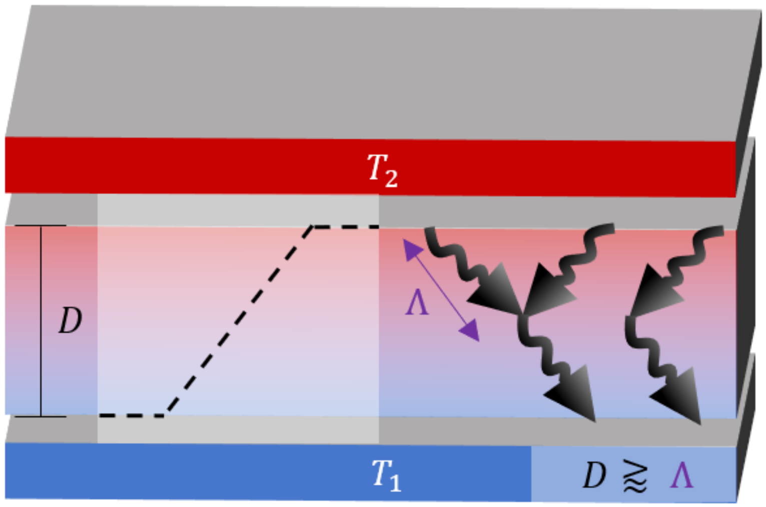
- Phonon mean free paths in bulk silicon are 10s of nm to microns

- Dimensions in nanoelectronics are much smaller




Phys. Rev B **84**, 085204 (2011)
Nature **530,** 144–147 (2016)

# Example: confinement

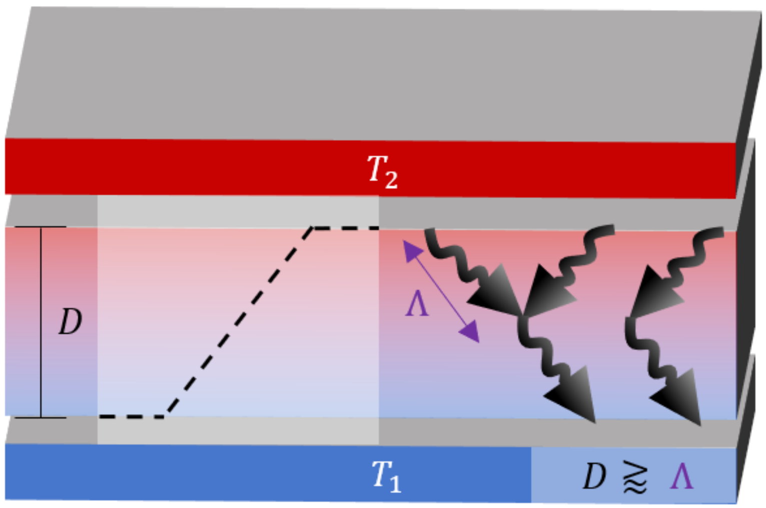



Heat flow across a film of similar thickness to the phonon mean free path

Naïve approach
"ballistic" model

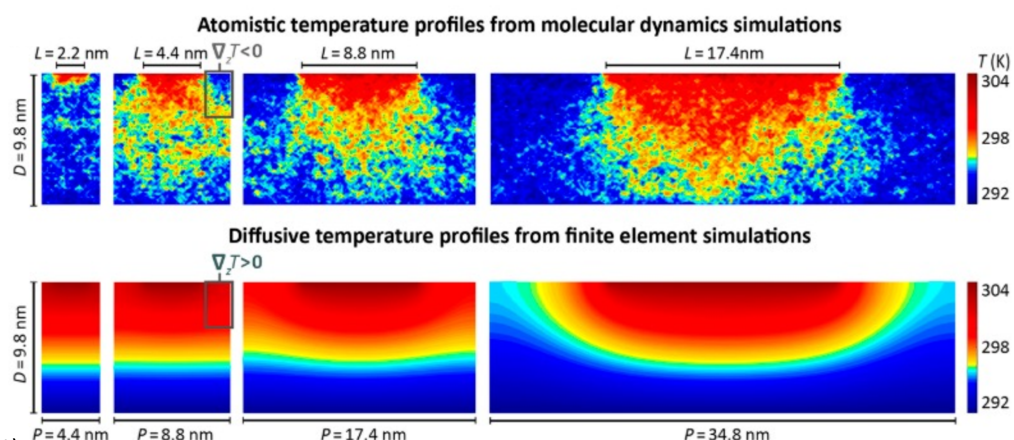
$$\kappa = \sum_k v_k \bar{\Lambda}_k c_k \quad \begin{cases} \bar{\Lambda}_k = D \text{ if } \Lambda_k > D \\ \bar{\Lambda}_k = \Lambda_k \text{ if } \Lambda_k < D \end{cases}$$

# Example: confinement




Heat flow across a film of similar thickness to the phonon mean free path

Sophisticated but (very) computationally intensive approach




PNAS **118** e2109056118 (2021)

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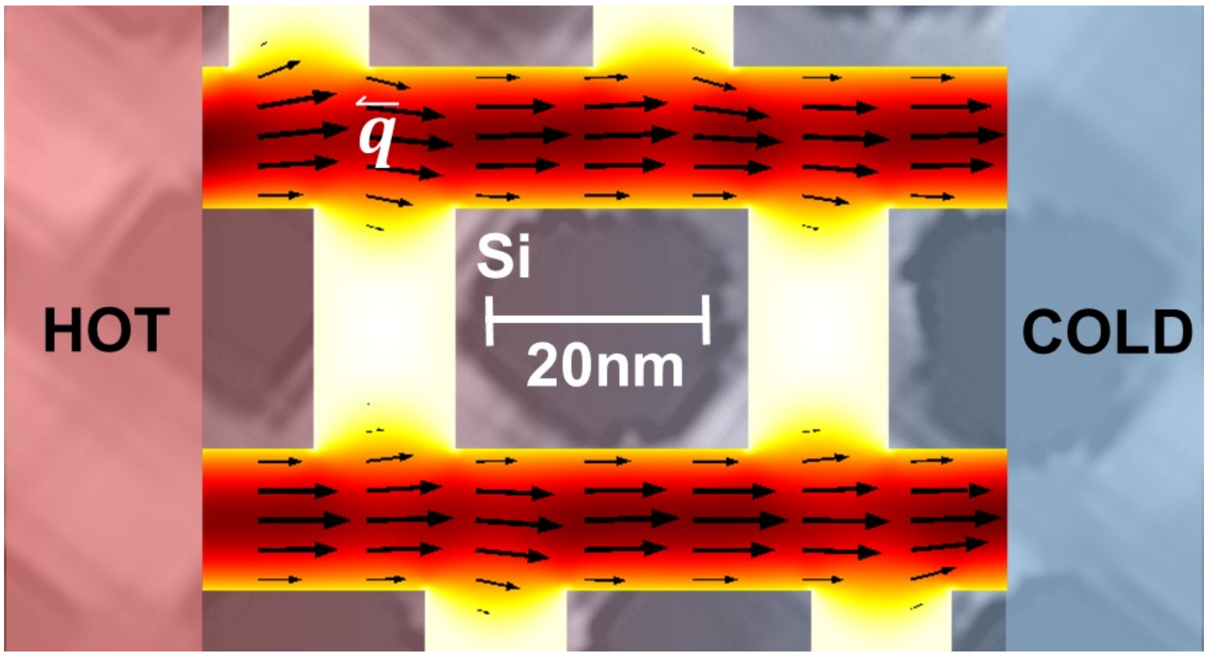



Nano Letters **23** 2129 (2023)

And what if your system looks like this?

Search for mesoscale theories: emergent phonon hydrodynamics

# Phonon hydrodynamics

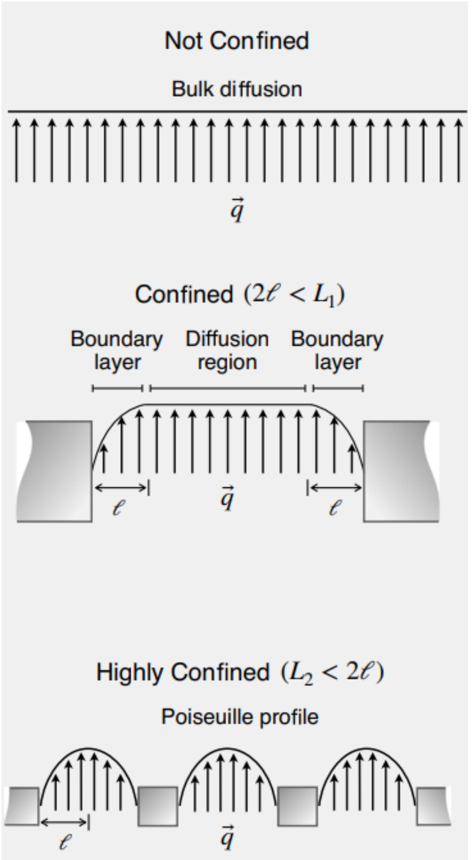
- Same process as before
- Thermodynamically-motivated distribution function at intermediate timescales

$$\mathbf{f}_\lambda = \mathbf{f}_\lambda^{\mathrm{eq}} + \vec{\beta}_\lambda \cdot \vec{\mathbf{q}} + \vec{\vec{\mathbf{G}}}_\lambda : \vec{\nabla}\vec{\mathbf{q}}$$

- Plug into the Boltzmann equation, arrive at an equation which looks like the Navier-Stokes Equation

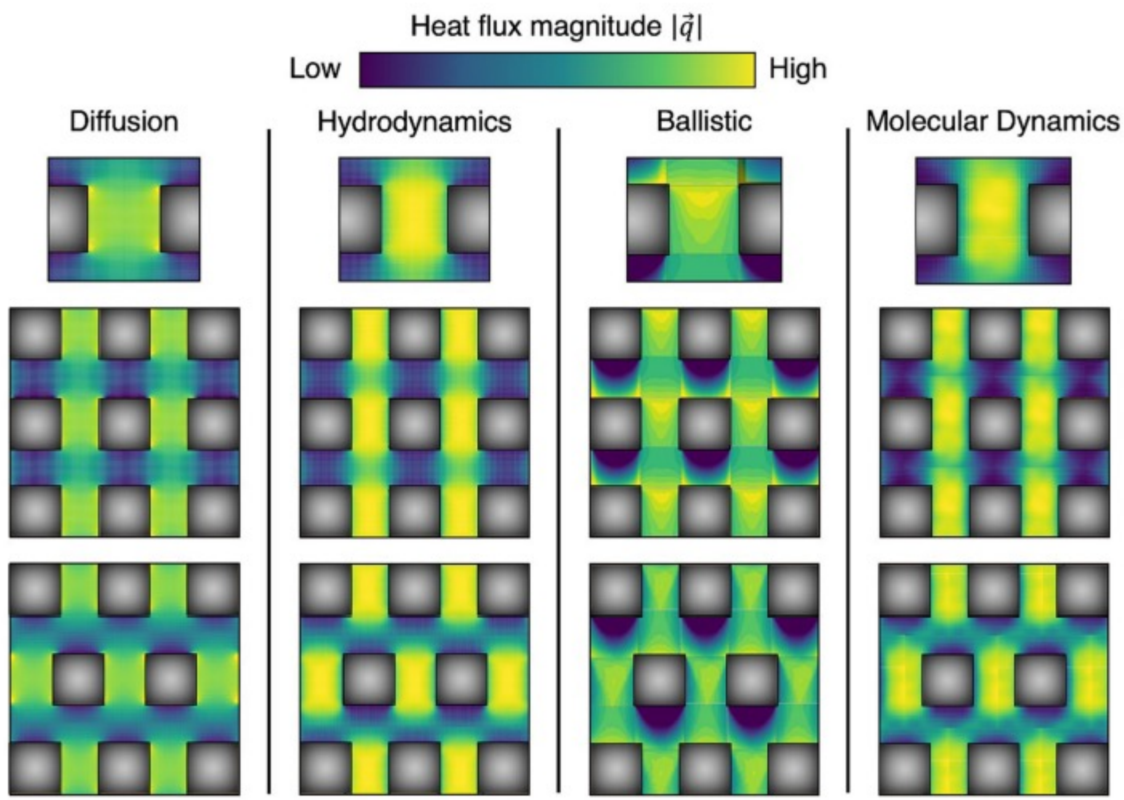
$$\vec{\mathbf{q}} = -\kappa_{\mathrm{GK}} \vec{\nabla}\mathbf{T} + \ell^2 \vec{\nabla}^2 \vec{\mathbf{q}}$$

viscous term




npj Comp. Mat. **11** 172 (2025)

# Phonon hydrodynamics (vs. other models)

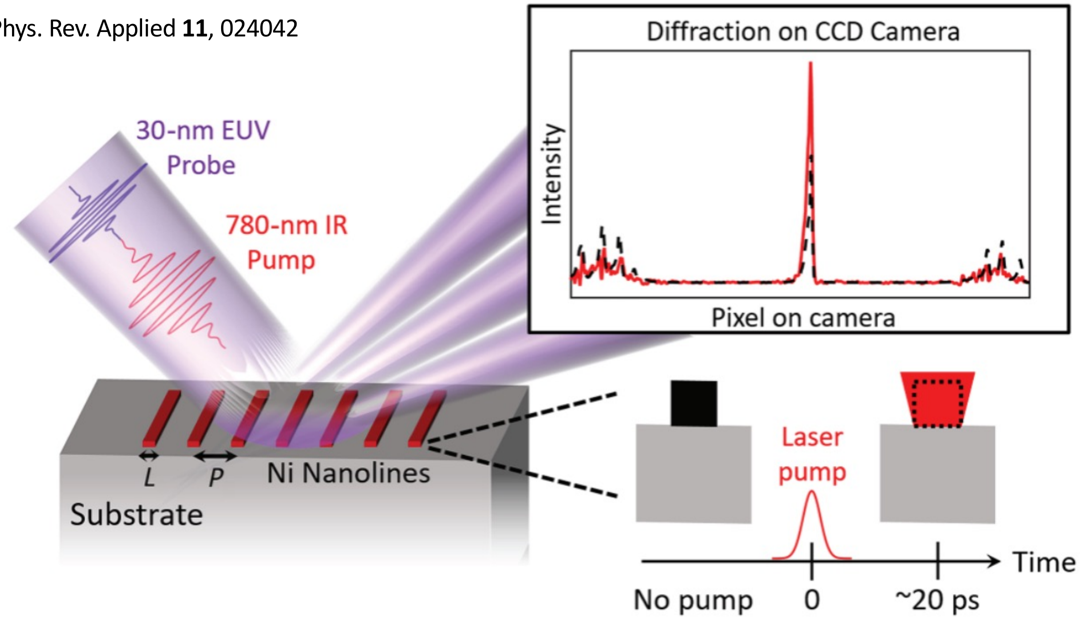



How can we develop experimental tools to differentiate between these models?

npj Comp. Mat. **11** 172 (2025)

# Example: EUV scatterometry

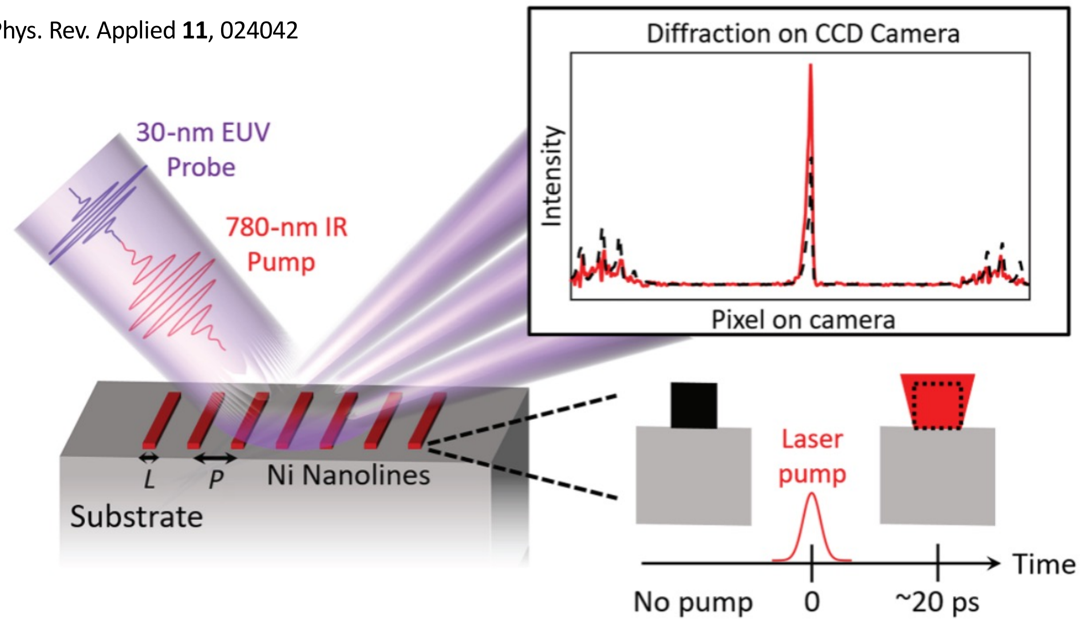
Phys. Rev. Applied **11**, 024042

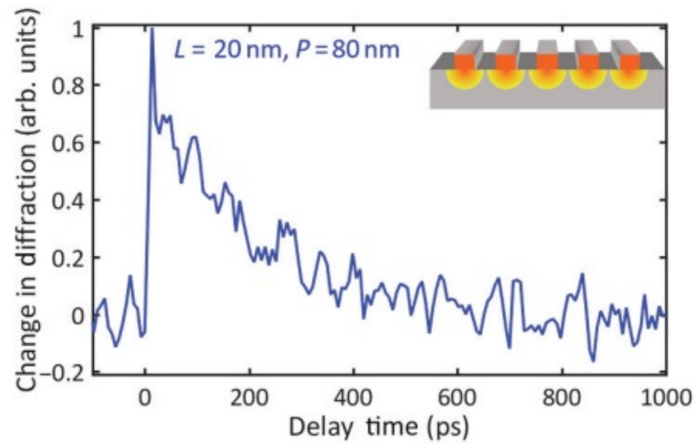


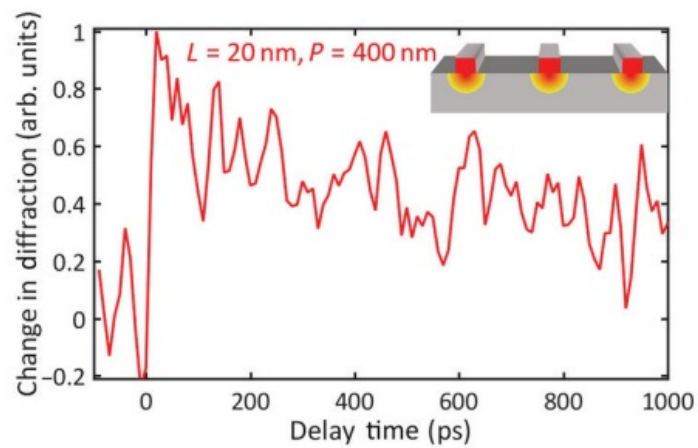

1. Heat Ni nanowires on a silicon substrate with an infrared laser
2. Measure dynamic diffraction pattern (thermal expansion and contraction) using ultrafast EUV light

# Example: EUV scatterometry

Phys. Rev. Applied **11**, 024042




1. Heat Ni nanowires on a silicon substrate with an infrared laser
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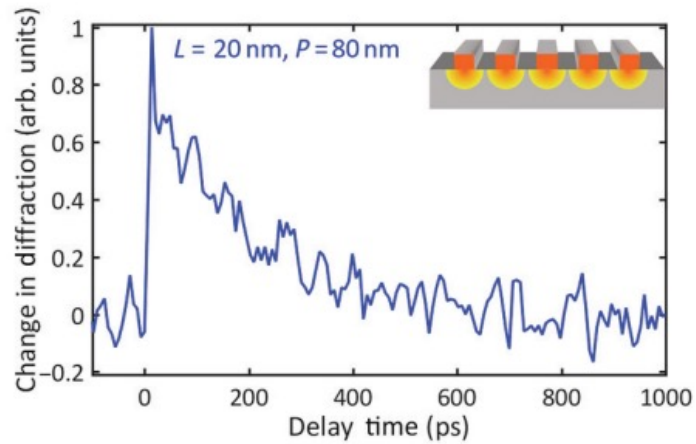
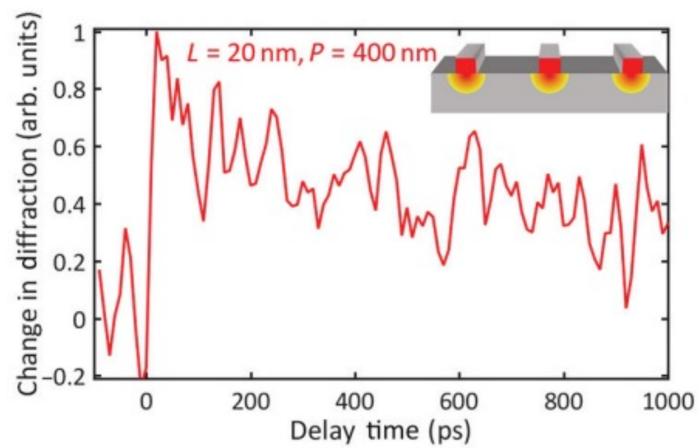
# Example: EUV scatterometry

Closely packed nanostructures cool faster than more isolated nanostructures under the same initial heat injection per unit volume!



# Summary

1. Phonons appear everywhere in condensed matter physics and their behavior is highly material/system dependent

2. Our knowledge of phonon transport is decades behind electron transport because phonons cannot be as easily manipulated

3. Phonon transport is critical for semiconductors, energy efficiency, quantum technologies and much more

# Thank you!