Heat conduction and phonon localization in disordered harmonic lattices

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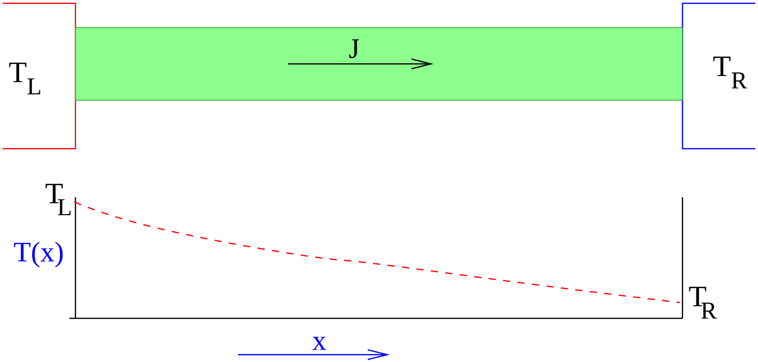
Outline

Heat CONDUCTION - Heat transfer through a body from HOT to COLD region.



Fourier's law

- Fourier's law of heat conduction

$$J = -\kappa \nabla T(x)$$

- Using Fourier's law and the energy conservation equation

$$\frac{\partial \epsilon}{\partial t} + \nabla . J = 0$$

gives the heat DIFFUSION equation:

$$\frac{\partial T}{\partial t} = \frac{\kappa}{c} \nabla^2 T$$

$c =$ specific heat capacity.

Fourier's law

For small $\Delta T = T_L - T_R$ and system size L :

$$\text{Fourier}'\text{s law implies :} \quad J \sim \kappa \frac{\Delta T}{L}$$

The thermal conductivity κ is an intrinsic material property.

e.g at room temperature:

$$\begin{aligned} \kappa &= 0.025\ W/mK \quad \text{air} \\ &= 429\ W/mK \quad \text{silver} \\ &= 2000\ W/mK \quad \text{Diamond} \end{aligned}$$

Proof of Fourier's law

Open problem:
Microscopic derivation/verification of Fourier's law in a realistic model with Hamiltonian dynamics.

Review article:
Fourier's law: A challenge for theorists
(Bonetto, Rey-Bellet, Lebowitz) (2000).

Results so far

Studies in one and two dimensional systems find that Fourier's law is not valid.

The thermal conductivity is not an intrinsic material property.

For anharmonic systems without disorder , κ diverges with system size L as:

$$
\begin{aligned}
\kappa &\sim L^{\alpha} \quad 1D \\
&\sim \log L \quad 2D
\end{aligned}
$$

Pinned anharmonic (momentum non-conserving) systems satisfy Fourier's law.

A.D, Advances in Physics, vol. 57 (2008).

Implications

This is interesting not just from the point of theory but also important from the experimental point of view.

The thermal conductivity of a 1 *mm* long carbon nanotube is $(1000)^{\alpha}$ times larger than that of a 1 μm long carbon nanotube !!

Recent experiments on nanotubes and graphene report diverging conductivity.

Higher dimensional systems

- What about systems in three dimensions ?
  Is Fourier's law valid ?
- Consider dielectric crystals.
  - Heat conduction is through lattice vibrations .
  - Harmonic approximation:
    solid can be thought of as a gas of phonons with frequencies corresponding to the vibrational spectrum of ordered harmonic crystal.
  - Phonons are the heat carriers. Phonons are scattered by:
    (i) Impurities
    (ii) Other phonons (anharmonicity)

    It is expected that thse scattering mechanisms should lead to a finite thermal conductivity κ.

Three dimensional systems

Look at what happens in the simplest three dimensional models.

- Heat conduction in a harmonic crystal with isotopic mass disorder (At low temperatures one can neglect anharmonicity) .
- Heat conduction in an anharmonic crystal.
- Look at L dependence of J in nonequilibrium steady state. ($J \sim 1/L$?)

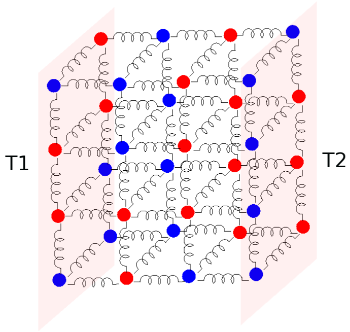
Heat conduction in mass disordered harmonic crystal.

Nearest neighbor harmonic Hamiltonian with scalar displacements.

$$H = \sum_{\mathbf{x}} \frac{m_{\mathbf{x}}}{2} \dot{z}_{\mathbf{x}}^2 + \sum_{\mathbf{x},\hat{\mathbf{e}}} \frac{k}{2} (z_{\mathbf{x}} - z_{\mathbf{x}+\hat{\mathbf{e}}})^2 + \sum_{\mathbf{x}} \frac{k_o}{2} z_{\mathbf{x}}^2$$

Masses $m_{\mathbf{x}}$ random.

$k_o = 0$: Unpinned. $k_o > 0$: Pinned.



Kinetic theory

Phonons do a random walk. A phonon of frequency ω has a mean free path $\ell_K(\omega)$. Thermal conductivity is given by:

$$\kappa = \int_0^{\omega_D} d\omega D(\omega)\rho(\omega)c(\omega, T)$$

$D(\omega) = v\ell_K(\omega) =$ phonon diffusivity ,
$v =$ phonon velocity ,
$\rho(\omega) =$ density of states ,
$c =$ specific heat capacity ,
$\omega_D =$ Debye frequency .

- Rayleigh scattering of phonons gives a mean free path $\ell_K(\omega) \sim 1/\omega^{d+1}$.
- For low frequency phonons $\ell_K \gtrsim L$, where L is size of system (ballistic heat transport).
- $\rho(\omega) \sim \omega^{d-1}$. Hence

$$\begin{aligned} \kappa &= \int_{1/L}^{\omega_D} d\omega \rho(\omega) v \ell_K(\omega) \\ &\sim L^{1/(d+1)} \end{aligned}$$

$1D \;\; \kappa \sim L^{1/2} : \; 2D \;\; \kappa \sim L^{1/3} : \; 3D \;\; \kappa \sim L^{1/4}$

Thus kinetic theory predicts a divergence of the conductivity in all dimensions.

Problems with kinetic theory

- Is a perturbative result valid in the limit of weak disorder. Rayleigh scattering result for $\ell_K(\omega)$ is obtained for single scattering of sound waves.

- Effect of Anderson localization is not accounted for since this requires one to consider multiple scattering of phonons.

- The kinetic theory result can also be derived directly from the Green-Kubo formula and $\ell_K(\omega)$ can be computed using normal mode eigenvalues and eigenstates.
  Probably correct for normal transport.
  Probably wrong for anomalous transport.

- Consider harmonic Hamitlonian:

$$H = \sum_{\mathbf{x}} \frac{1}{2} \dot{Z}_{\mathbf{x}} M_{\mathbf{x}\mathbf{x}} \dot{Z}_{\mathbf{x}} + \sum_{\mathbf{x},\mathbf{x}'} \frac{1}{2} Z_{\mathbf{x}} \phi_{\mathbf{x}\mathbf{x}'} Z_{\mathbf{x}'} \ .$$

x denotes point on a d-dimensional hypercube of size N^d.
M- Mass-matrix, ϕ- Force-matrix.

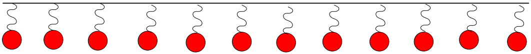
- Equation for q^{th} normal mode, $q = 1, 2, ..., N^d$.

$$m_{\mathbf{x}} \ \Omega_q^2 \ a_q(\mathbf{x}) = \sum_{\mathbf{x}'} \ \phi_{\mathbf{x}\mathbf{x}'} \ a_q(\mathbf{x}') \ .$$

Normal mode frequency: Ω_q
Normal mode wave-function: $a_q(\mathbf{x})$

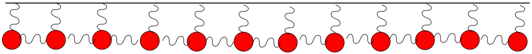
Normal modes

Consider N uncoupled oscillators.



N localized normal modes each with $\Omega_q = (k_o/m)^{1/2}$.

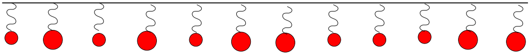
Now couple the oscillators by springs with $k << k_o$.



We get N extended plane wave states with:

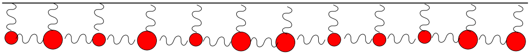
$$
\begin{aligned}
a_q(l) &= \sin k_q l \\
\Omega_q^2 &= \left(\frac{k_o}{m} + \frac{4k}{m} \sin^2 k_q/2 \right) \\
k_q &= q\pi/(N+1) \,.
\end{aligned}
$$

Consider N uncoupled oscillators with random masses (m_1, m_2).



N localized normal modes with $\Omega_q = (k_o/m_1)^{1/2}, (k_o/m_2)^{1/2}$.

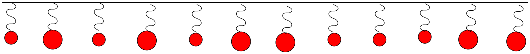
Now couple the oscillators by springs with $k << k_o$.



Degenerate states of strings of equal masses linearly combine to form new states.

All normal modes are localized.

Consider N uncoupled oscillators with random masses (m_1, m_2).



N localized normal modes with $\Omega_q = (k_o/m_1)^{1/2}, (k_o/m_2)^{1/2}$.

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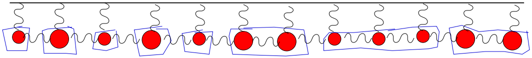
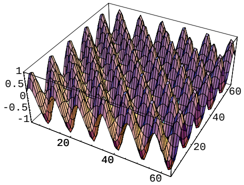


Degenerate states of strings of equal masses linearly combine to form new states.

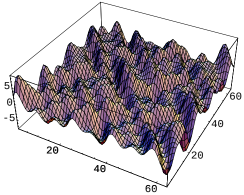
All normal modes are localized.

Character of normal modes

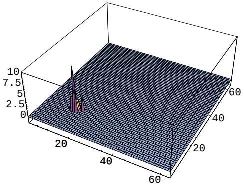
In general normal modes can be of three types:



Extended periodic mode
(Ballistic)



Extended random mode
(Diffusive)



Localized Mode
(Non-conducting)

Predictions from Renormalization group theory

S. John, H. Sompolinsky, and M.J. Stephen

For unpinned system:

- In $1D$ all modes with $\omega > 1/N^{1/2}$ are localized.
- In $2D$ all modes with $\omega > [\log(N)]^{-1/2}$ are localized.
- In $3D$ there is a band of low frequency non-localized states between $0 - \omega_c^L$ where ω_c^L is independent of system-size.

System size dependence of current ??
Nature of non-localized states ?? -
Ballistic OR Diffusive OR .....

Nature of phonon modes

Localization theory tells us that low frequency modes are extended.

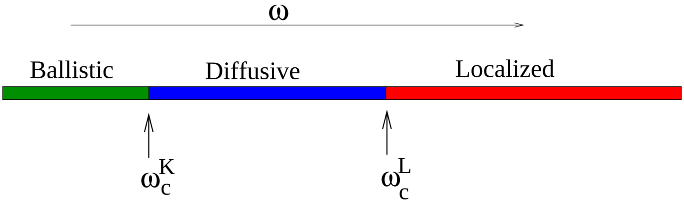
At low frequencies the effect of disorder is weak and so expect kinetic theory to be accurate.

Setting $\ell_K(\omega) = N$ gives us the frequency scale ω_c^K below which modes are ballistic. Since $\ell_K(\omega) = 1/\omega^{d+1}$ this gives

- $1D: \quad \omega_c^K \sim N^{-1/2}$
- $2D: \quad \omega_c^K \sim N^{-1/3}$
- $2D: \quad \omega_c^K \sim N^{-1/4}$

Nature of phonon modes for unpinned system

Kinetic theory and localization theory gives us two frequency cut-offs: ω_c^K and ω_c^L:



- $1D: \quad \omega_c^K \sim \omega_c^L \sim N^{-1/2}$
- $2D: \quad \omega_c^K \sim N^{-1/3}, \quad \omega_c^L \sim [\ln N]^{-1/2}$
- $3D: \quad \omega_c^K \sim N^{-1/4}, \quad \omega_c^L \sim N^0$

Try to estimate the N dependence of J.

Heuristic theory

- Nonequilibrium heat current is given by the Landauer formula:

$$J = \frac{\Delta T}{2\pi} \int_0^\infty d\omega \mathcal{T}(\omega) \ ,$$

  where $\mathcal{T}(\omega)$ is the transmission coefficient of phonons.

- Phonons can be classified as:

  Ballistic: Plane wave like states with $\mathcal{T}(\omega)$ independent of system size.

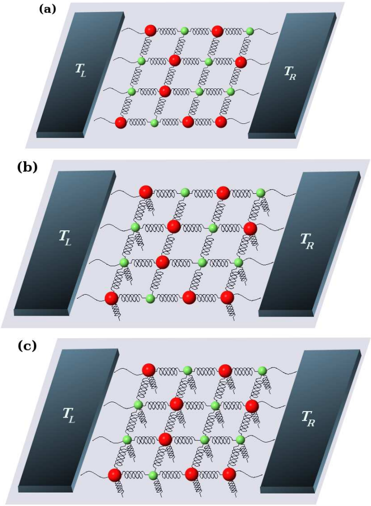
  Diffusive: Extended disordered states with transmission decaying as $\mathcal{T}(\omega) \sim 1/N$.

  Localized states: $\mathcal{T}(\omega) \sim e^{-N/\ell_L}$.

- Find contribution to J of different types of modes and estimate the asymptotic size dependence.

- Ballistic contribution to current depends on boundary conditions.

Boundary conditions



Free BC: $\mathcal{T}(\omega) \sim \omega^{d-1}$
$\omega \to 0$

Fixed BC: $\mathcal{T}(\omega) \sim \omega^{d+1}$
$\omega \to 0$

Pinned system: No low-frequency ballistic modes.

Analytical predictions of heuristic theory

1D unpinned fixed BC: $J \sim N^{-3/2}$
1D unpinned free BC: $J \sim N^{-1/2}$
1D pinned: $J \sim exp(-bN)$

These agree with rigorous results.

2D unpinned fixed BC: $J \sim N^{-1}(\ln N)^{-1/2}$

2D unpinned free BC: $J \sim N^{-2/3}$

2D pinned: $J \sim exp(-bN)$

3D unpinned fixed BC: $J \sim N^{-1}$

3D unpinned free BC: $J \sim N^{-3/4}$

3D pinned: $J \sim N^{-1}$

We now check these predictions numerically.

Disordered harmonic crystal driven by Langevin heat baths at different temperatures.

Equations of motion:

$$m_{\mathbf{x}} \ddot{z}_{\mathbf{x}} = -\sum_{\hat{\mathbf{e}}} (z_{\mathbf{x}} - z_{\mathbf{x}-\hat{\mathbf{e}}}) - k_o z_{\mathbf{x}} \quad \{+f_{\mathbf{x}} \text{ from heat baths } \}$$

$\{m_{\mathbf{x}}\}$ chosen from a random distribution.

$f_{\mathbf{x}}$ is force from heat baths and acts only on boundary particles.
We choose white noise Langevin baths:

$$\begin{aligned} f_{\mathbf{x}} &= -\gamma \dot{z}_{\mathbf{x}} + (2\gamma T)^{1/2} \eta_{\mathbf{x}}(t) \\ \langle \eta_{\mathbf{x}}(t) \eta_{\mathbf{x}'}(t') \rangle &= \delta(t - t') \delta_{\mathbf{x},\mathbf{x}'} \end{aligned}$$

Periodic boundary conditions in transverse directions.

Measured quantities

In the nonequilibrium steady state we measure the energy current and temperature profile. These are given by the following time-averages ($1D$):

$$
\begin{aligned}
J &= \frac{1}{N-1}\sum_{l=2}^{N}\langle z_{l-1}\dot{z}_l\rangle \,, \\
T_l &= m_l\langle \dot{z}_l^2\rangle \,.
\end{aligned}
$$

We use two approaches:

1. Numerical evaluation of phonon transmission and using the Landauer formula to find J.

2. Nonequilibrium Simulations to find J.

The Landauer formula

- Linear equations: Can be solved by Fourier transforms.

$$
\begin{aligned}
\tilde{z}(\omega) &= G(\omega)\tilde{\eta}(\omega) \\
G(\omega) &= [-M\omega^2 + \phi - \Sigma(\omega)]^{-1}
\end{aligned}
$$

- Steady state quantities of interest are expectation values of quadratics and can be evaluated using properties of noise correlations.
- Final result for current can be expressed in terms of the transmission coefficient $\mathcal{T}(\omega)$.

  This gives the Landauer formula.

  $\mathcal{T}(\omega)$ can be written in terms of the phonon Green's functions $G(\omega)$.

$$J = \frac{\Delta T}{2\pi} \int_0^\infty d\omega \mathcal{T}(\omega) \; ,$$

where

$$\mathcal{T}(\omega) = 4\gamma^2 \omega^2 |G_{1N}|^2 \; ,$$

and

$$\begin{aligned} G &= [-\omega^2 M + \Phi - \Sigma]^{-1} \\ &= \begin{pmatrix} a_1 - i\gamma\omega & -1 & 0 & \ldots & 0 \\ -1 & a_2 & -I & 0 \ldots & 0 \\ \ldots & \ldots & \ldots & \ldots & \ldots \\ 0 & \ldots 0 & -1 & a_{N-1} & -1 \\ 0 & \ldots & 0 & -1 & a_N - i\gamma\omega \end{pmatrix}^{-1} \; , \end{aligned}$$

$a_l = 2 + k_o - m_l \omega^2$.

Product of random matrices

Transmission function $\mathcal{T}(\omega)$ can be expressed in terms of a product of random matrices.

$$\text{Then } G_{1N} = (1 \quad -i\gamma)\ \hat{T}_N\ \hat{T}_{N-1} \ldots \hat{T}_1 \begin{pmatrix} 1 \\ i\gamma \end{pmatrix}$$

$$\text{where } \hat{T}_l = \begin{pmatrix} 2 - m_l\omega^2 & -1 \\ 1 & 0 \end{pmatrix}$$

Generalization to higher dimensions (N^d lattice):
$\mathcal{T}(\omega)$ can be expressed in terms of a product of $2N^{d-1} \times 2N^{d-1}$ random matrices (Anupam Kundu).

We implement this numerically for $d = 2, 3$.

Numerical and simulation results

Open system. We look at:

- Disorder averaged transmission: $T(\omega) = \frac{[\mathcal{T}(\omega)]}{N^{d-1}}$
- Current: $J \sim \int_0^\infty d\omega \; T(\omega)$.

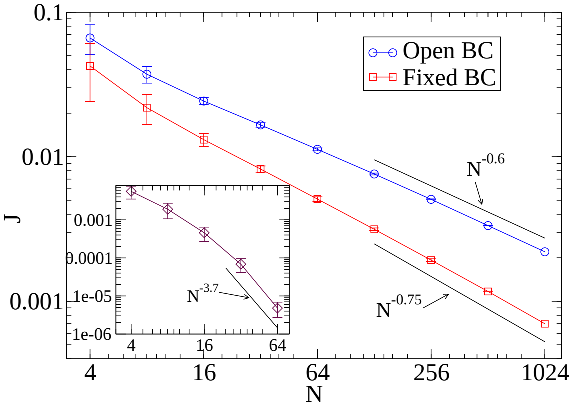
Closed system without heat baths.
For the normal modes find:

- Density of states: $\rho(\omega) = \frac{1}{N^d} \sum_q \delta(\omega - \Omega_q)$
- Inverse participation ratio: $P^{-1} = \frac{\sum_{\mathbf{x}} a_{\mathbf{x}}^4}{(\sum_{\mathbf{x}} a_{\mathbf{x}}^2)^2}$.

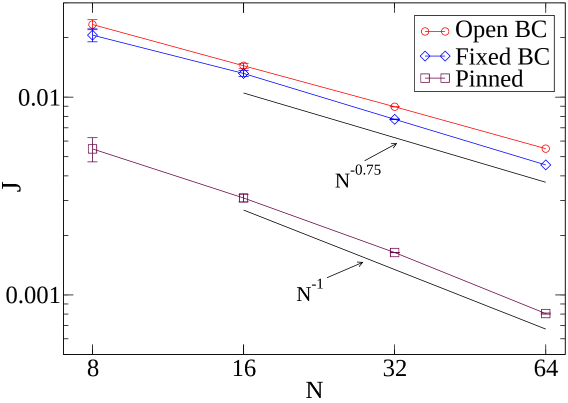
IPR is large (~ 1) for localized states, small ($\sim N^{-d}$) for extended states.

Binary mass distribution: $m_1 = 1 + \Delta$, $m_2 = 1 - \Delta$ $\quad\quad \Delta$ = Disorder strength.

J-versus-N plots: 2D



J-versus-N plots: 3D

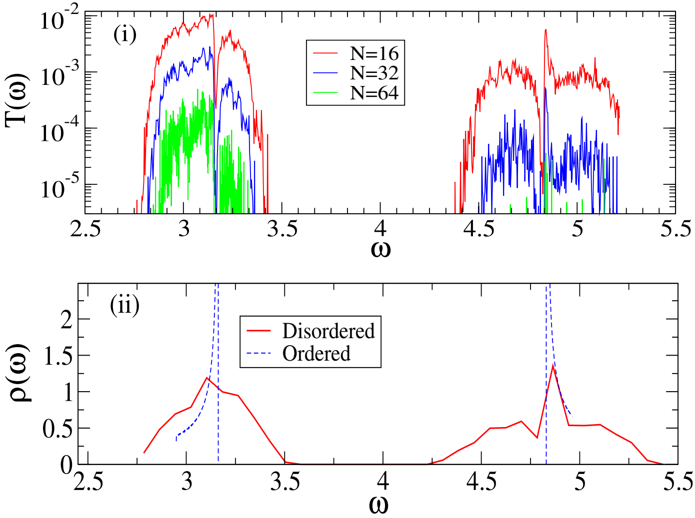


Comparision with numerical results

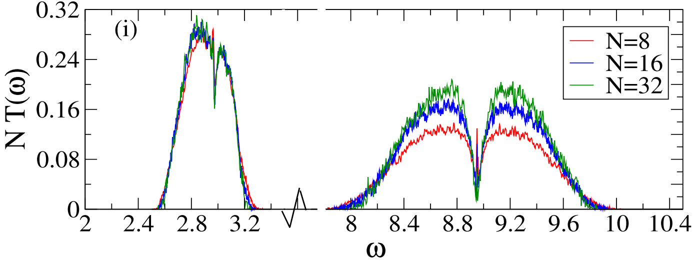
	Analytic prediction	(Numerical result)
2D unpinned fixed BC:	$J \sim N^{-1}(\ln N)^{-1/2}$	$[\sim N^{-0.75}]$
2D unpinned free BC:	$J \sim N^{-2/3}$	$[\sim N^{-0.6}]$
2D pinned:	$J \sim exp(-bN)$	$[\sim N^{-3.7}]$
3D unpinned fixed BC:	$J \sim N^{-1}$	$[\sim N^{-0.75}]$
3D unpinned free BC:	$J \sim N^{-3/4}$	$[\sim N^{-0.75}]$
3D pinned:	$J \sim N^{-1}$	$[\sim N^{-1}]$

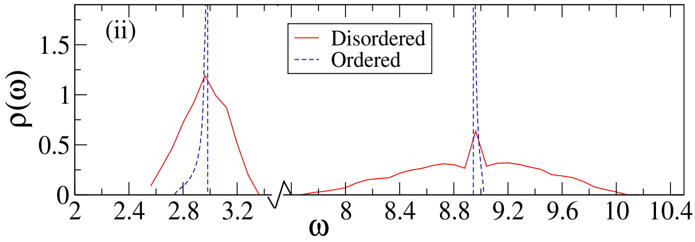
Look at Phonon transmission $T(\omega)$.

Phonon transmission: pinned $2D$

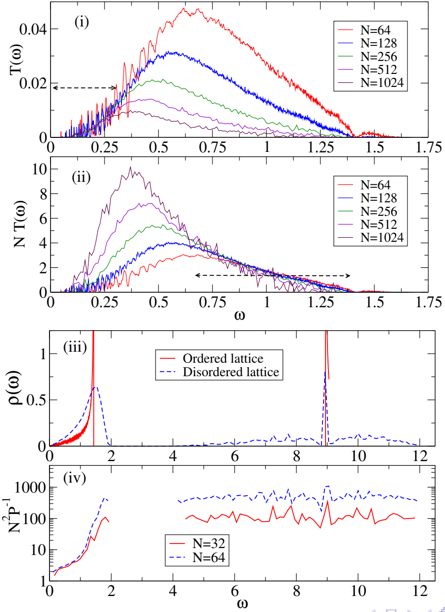


Phonon transmission: pinned $3D$

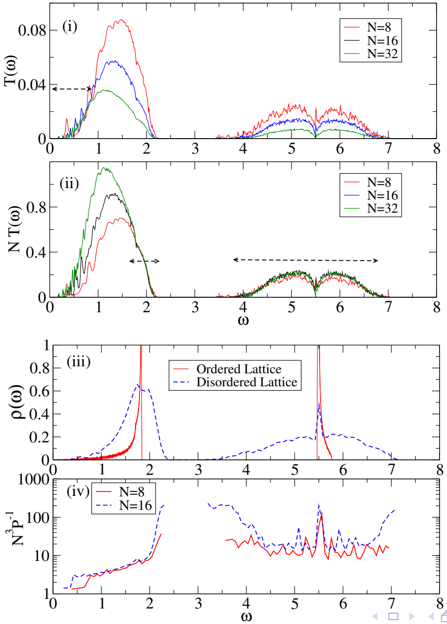




Transmission: Unpinned 2*D*



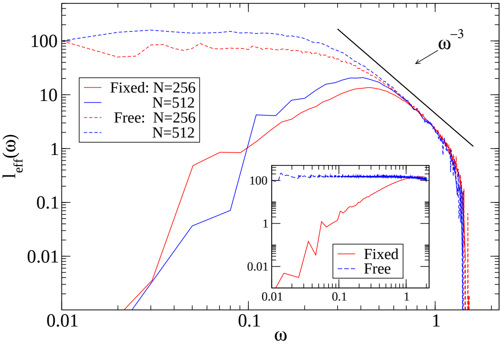
Transmission: Unpinned $3D$



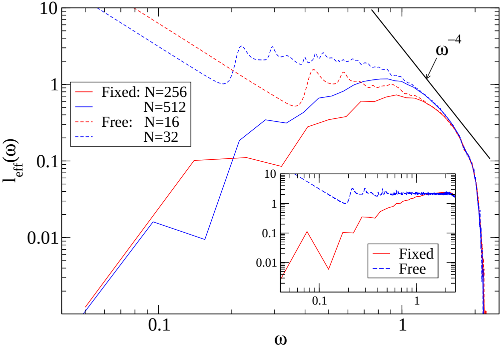
Fixed and Free BCs: $2D$

Define effective mean free path: l_{eff} through:

$NT(\omega)/2\pi = v(\omega) l_{\text{eff}}(\omega) \rho(\omega)$



Fixed and Free BCs: $3D$



Conclusions

- Unpinned system:
  - Results sensitive to boundary conditions.
  - Fourier's law valid for generic boundary conditions (fixed BC).
  - Numerical verification requires larger system sizes.
- Pinned system (closer to the electron localization problem):
  - First numerical verification of Fourier's law in a $3D$ system.

    Absence of transtion to insulating phase on increasing disorder.
  - Direct verification of a $2D$ heat insulator.

Future questions

- Analytic results
- Improving numerical methods and study larger system sizes.
- Absence of conductor-insulator transition: Connection to percolation ?
- Landauer and Green-Kubo formulas.

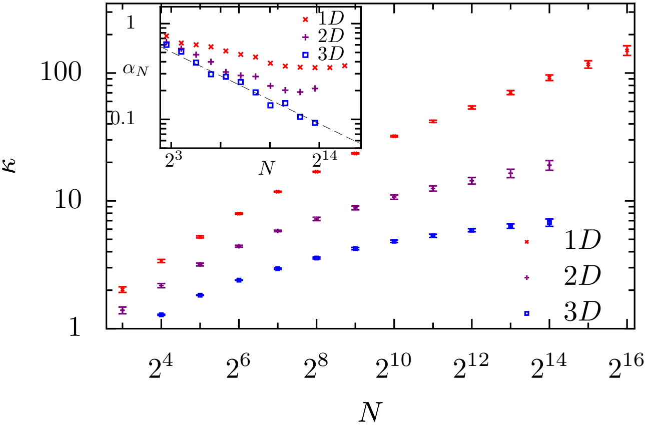
  $NT(\omega)/2\pi ? \ = ?D(\omega)\rho(\omega)$ (PROOF)

  In systems with normal and anomalous transport.
- What happens in real systems ? Effect of anharmonicity, other degrees of freedom.

  [Heat transport in model jammed solids: Nagel *etal*]

Heat Conduction in a Three Dimensional Ordered Anharmonic Crystal

Fermi-Pasta-Ulam interactions. Interparticle potential= x^4.



AD and Keiji Saito ——PRL **104** 040601 (2010).

Phonon conduction: differences with electron conduction.

- Phonons at all frequencies participate in heat transport. In electron case only electrons near Fermi-level contribute to electrical current.
- In phonon case, low-frequency modes are unaffected by disorder and are the main carriers of heat.