Ordered and Disordered Contributions to Lattice Thermal Conductivity

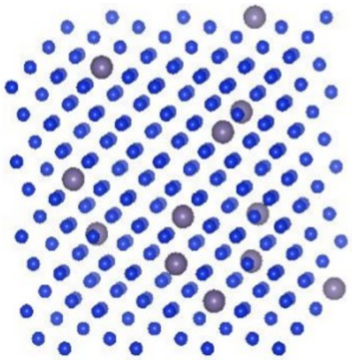
Jason Larkin and Alan McGaughey
Nanoscale Transport Phenomena Laboratory
Carnegie Mellon Department of Mechanical Engineering
http://ntpl.me.cmu.edu/

07/12/2012

Mechanical Engineering

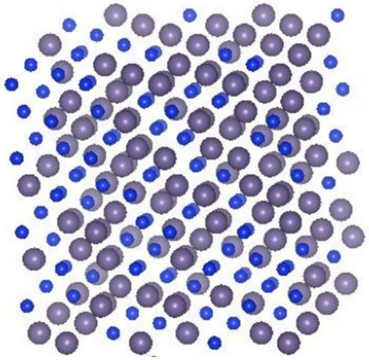
Thermal Transport in Disordered Materials

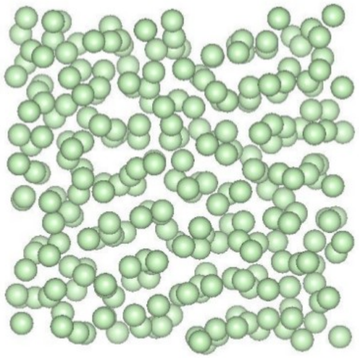
- **Phonon** picture valid with perturbations. **Perfect systems and dilute alloys**



- Phonon picture only valid for very long wavelengths. **Alloy and amorphous**

- **Localized** vibrations (diffusons) become important.





$$k_{vib} = k_{AF} + k_{ph}$$

(disorder) (order)

Mechanical Engineering

Theoretical Models

$$k_{vib} = k_{AF} + k_{ph}$$

Phonons (ordered): Interacting Phonon Gas

$$k_{ph} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} C_{ph}\binom{\boldsymbol{\kappa}}{\nu}\, v^2_{g,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu}\, \tau\binom{\boldsymbol{\kappa}}{\nu}$$

$$\boldsymbol{v}_g = \partial\omega/\partial\boldsymbol{\kappa}$$

$$\Lambda\binom{\boldsymbol{\kappa}}{\nu} = |\boldsymbol{v}_g|\tau\binom{\boldsymbol{\kappa}}{\nu}$$

- phonon-phonon (anharmonicity), defects (dilute), boundaries, (phonon-diffuson?)

Diffusons (disordered): Allen-Feldman Theory

$$k_{AF} = \sum_i C(\omega_i) D_{AF}(\omega_i)$$

$\boldsymbol{v}_g =$?

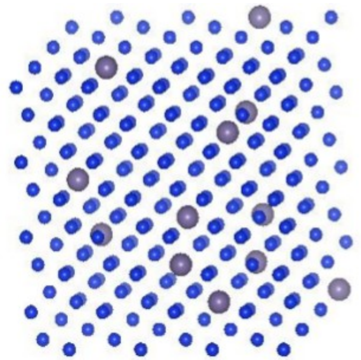
$\Lambda =$?

- diffuson-diffuson, (boundaries, phonon-diffuson?)

$$C_{ph}\binom{\boldsymbol{\kappa}}{\nu} = k_B/\mathrm{V} \qquad C(\omega_i) = k_B/\mathrm{V}$$

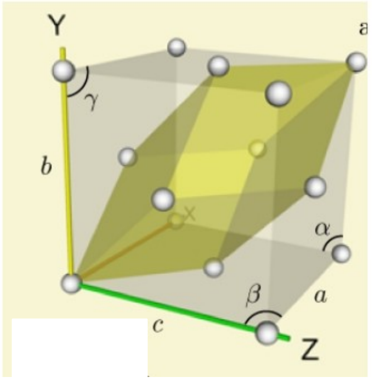
Mechanical Engineering

Modeled Systems: LJ Alloys and Amorphous

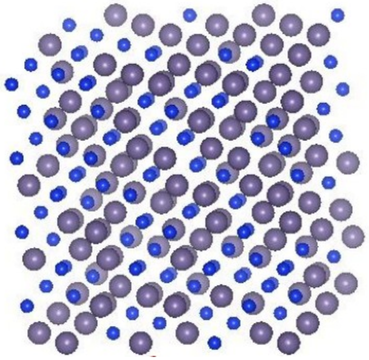


Virtual Crystal (VC)

$$\tau\binom{\boldsymbol{\kappa}}{\nu}$$



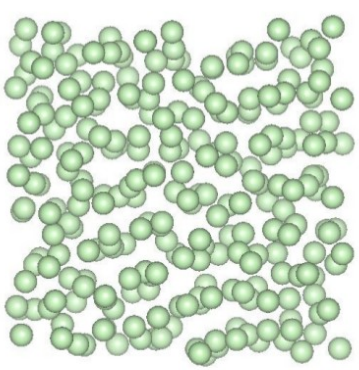
Unit cell



$$m^{a}_{1-c}m^{b}_{c} \quad m^{a}=1 \quad m^{b}=3$$

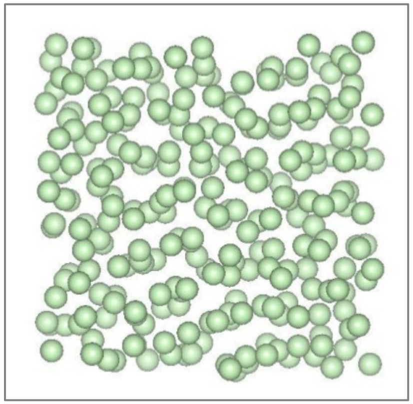
$$c=0.5,\ m_{avg}=2.0$$

Gamma point



$$\tau\binom{\boldsymbol{\kappa}=0}{\omega}$$

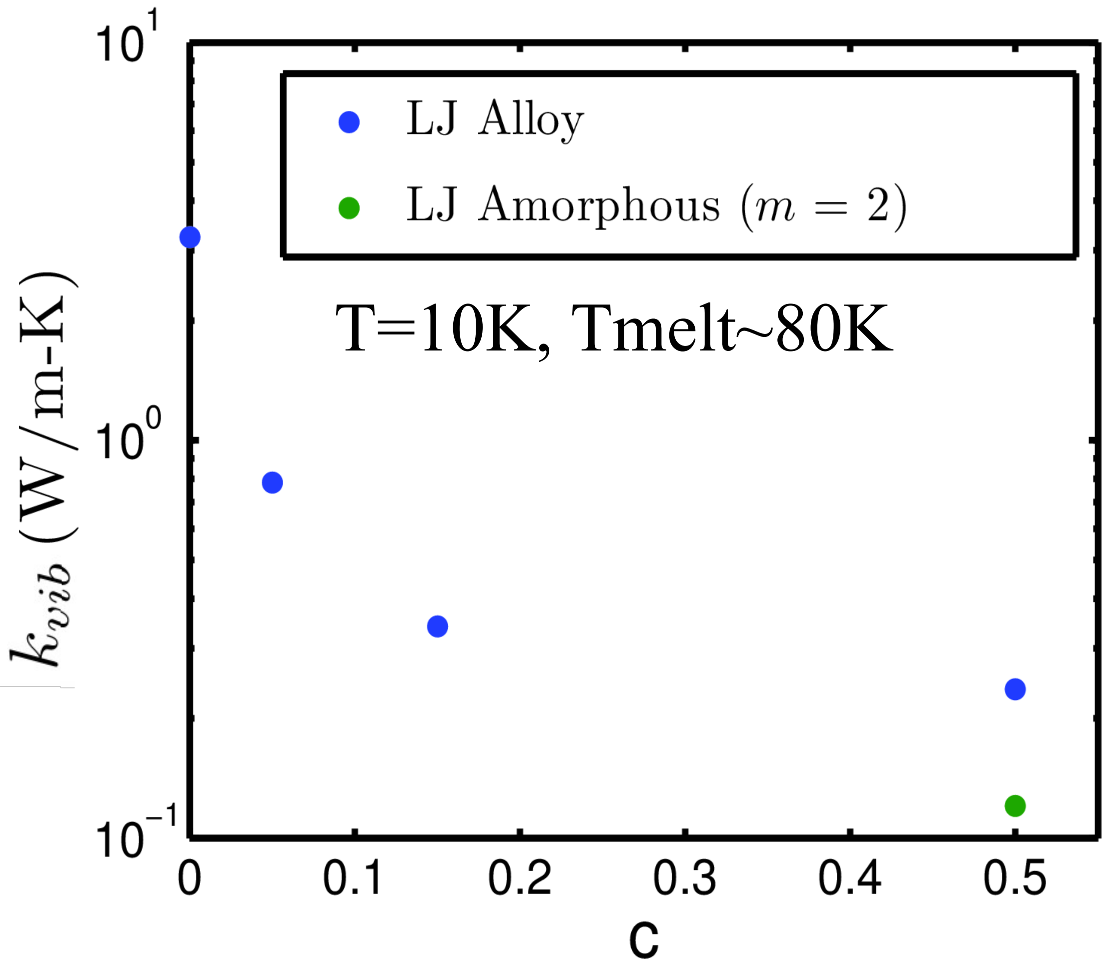
Unit cell



Thermal Conductivity: System-Level

Green-Kubo

$$k_{vib} = \frac{V}{3k_B T^2} \int_0^\infty \langle \mathbf{J}(0) \cdot \mathbf{J}(t) \rangle \, dt$$



- Heat current J has all effects of MD (anharmonicity, defects, etc.)

Mechanical Engineering

Normal Mode Decomposition (NMD)

$$q(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix};t) = \sum_{\alpha,b,l}^{3,n,N} \sqrt{\frac{m_b}{N}}\, u_\alpha(\begin{smallmatrix}l\\ b\end{smallmatrix};t)\, e^*(\begin{smallmatrix}\boldsymbol{\kappa} & b\\ \nu & \alpha\end{smallmatrix}) \exp[i\boldsymbol{\kappa}\cdot\mathbf{r}_0(\begin{smallmatrix}l\\ 0\end{smallmatrix})]$$

Atomic pos/vel from MD (anharmonicity, defects, etc)

Eigenvectors (HLD) w/ VC or Gamma

Allowed or Gamma point

Phonon/diffuson lifetimes:

$$\exp\left[-t/\tau\begin{pmatrix}\boldsymbol{\kappa}\\ \nu\end{pmatrix}\right] = \frac{\langle E_{\boldsymbol{\kappa},\nu}(t)E_{\boldsymbol{\kappa},\nu}(0)\rangle}{\langle E_{\boldsymbol{\kappa},\nu}(0)E_{\boldsymbol{\kappa},\nu}(0)\rangle}$$

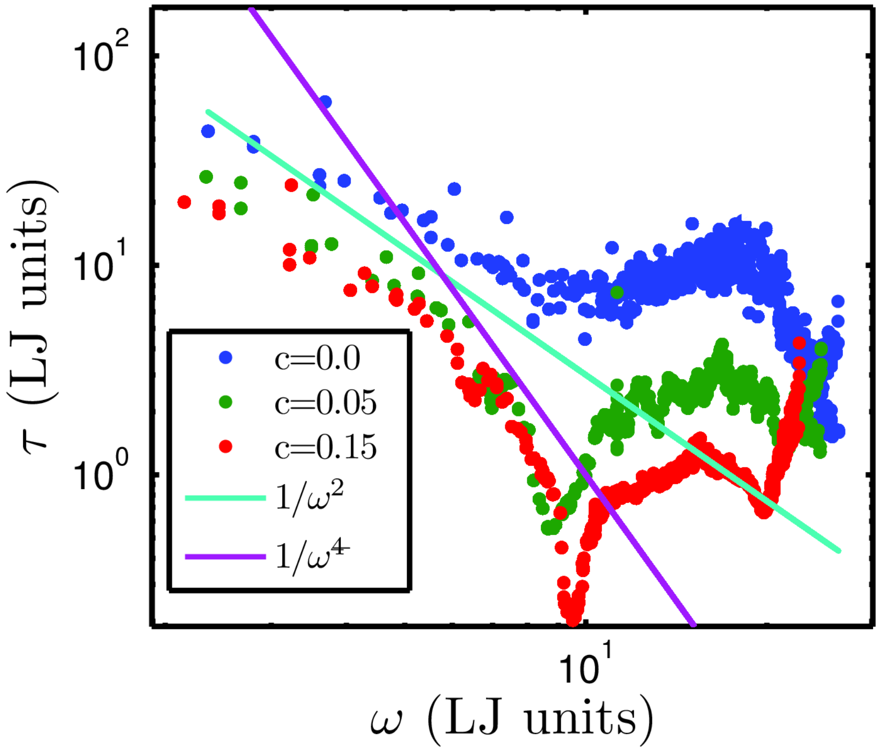
- Includes all effects of MD.
- Limited by HLD mapping.

PHYSICAL REVIEW B 79, 064301 (2009)

Mechanical Engineering

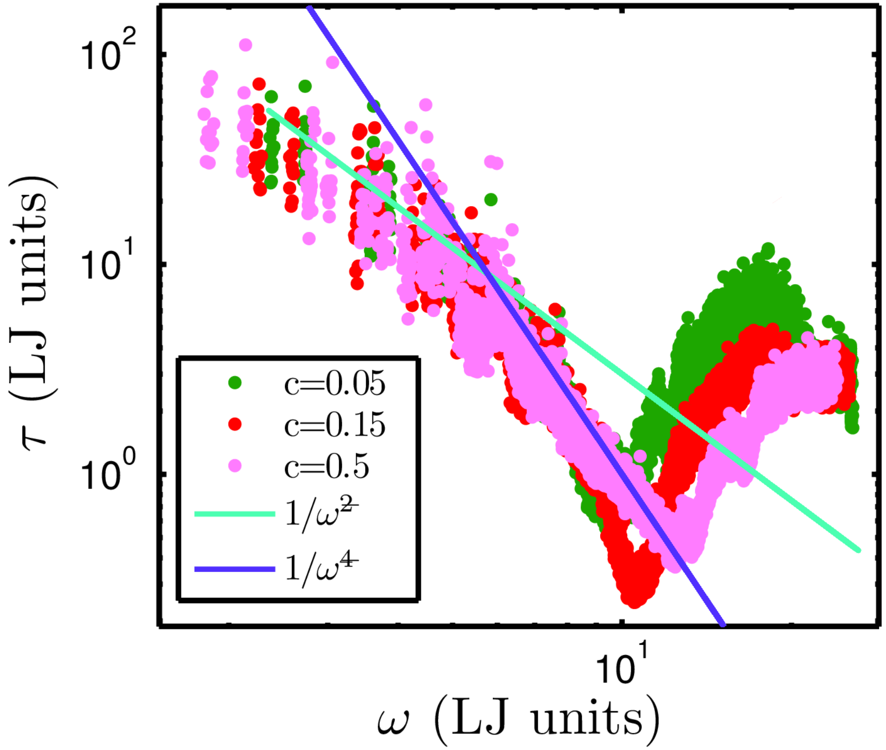
Normal Mode Decomposition (NMD)

Virtual Crystal



$$\tau\binom{\boldsymbol{\kappa}}{\nu}$$

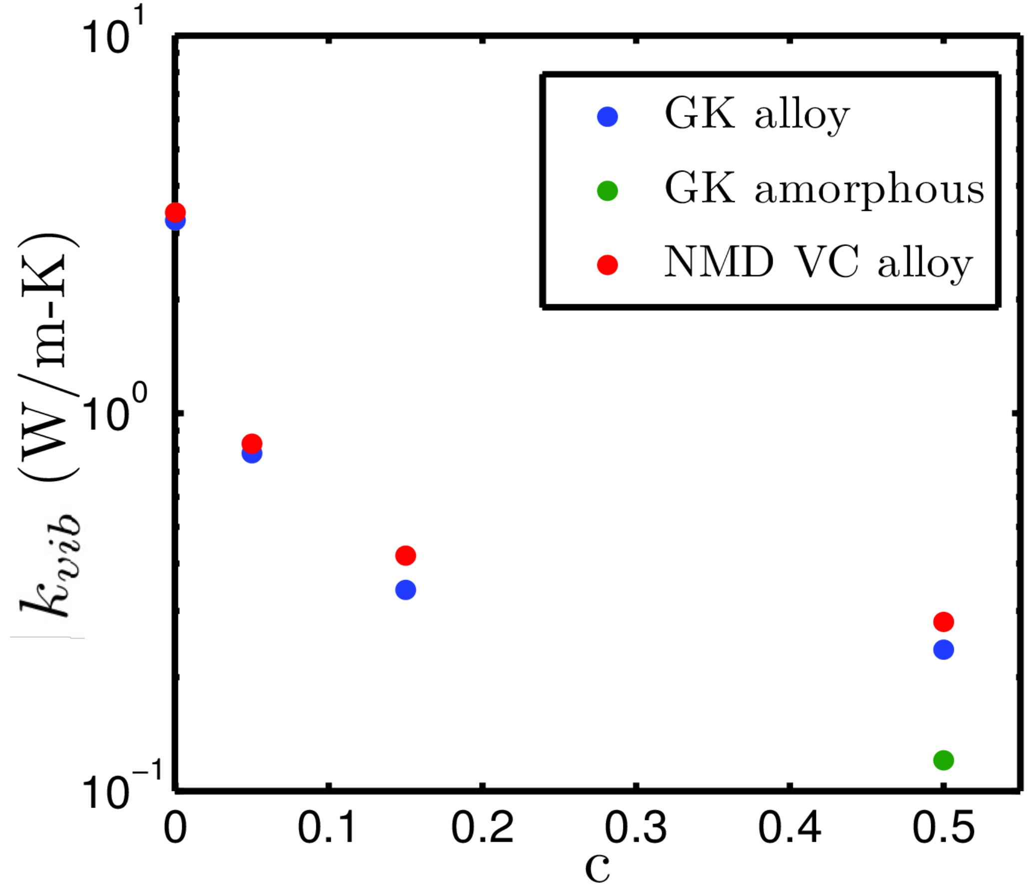
Gamma point



$$\tau\binom{\boldsymbol{\kappa} = 0}{\omega}$$

Thermal Conductivity: System- and Carrier-Level

Virtual Crystal approximation and phonon scaling relations work well!



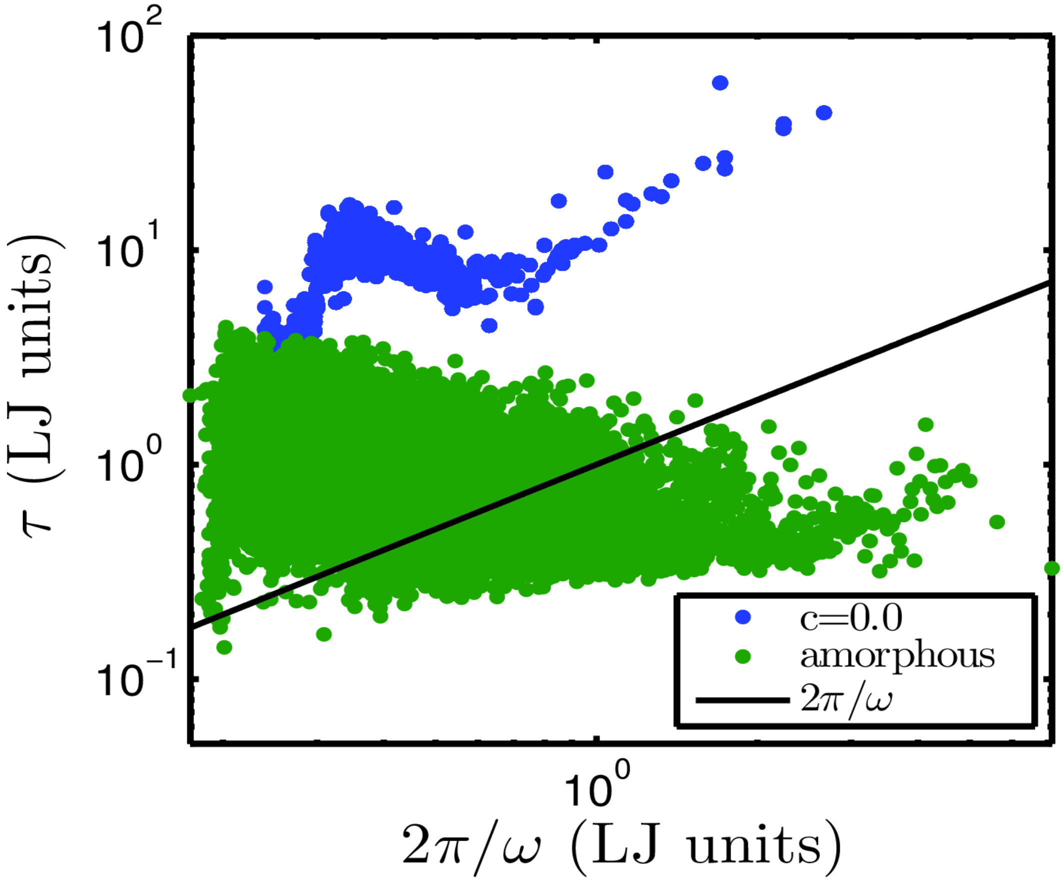
Anharmonic Lattice Dynamics + Defect scaling =

PHYSICAL REVIEW B 85, 184303 (2012)

PRL 106, 045901 (2011)

Mechanical Engineering

Ordered and Disordered Vibrations



Ioffe-Regel Limit:

$$\tau(\omega) = 2\pi/\omega$$

PHIL. MAG. B 79, 1715-1731 (1999)

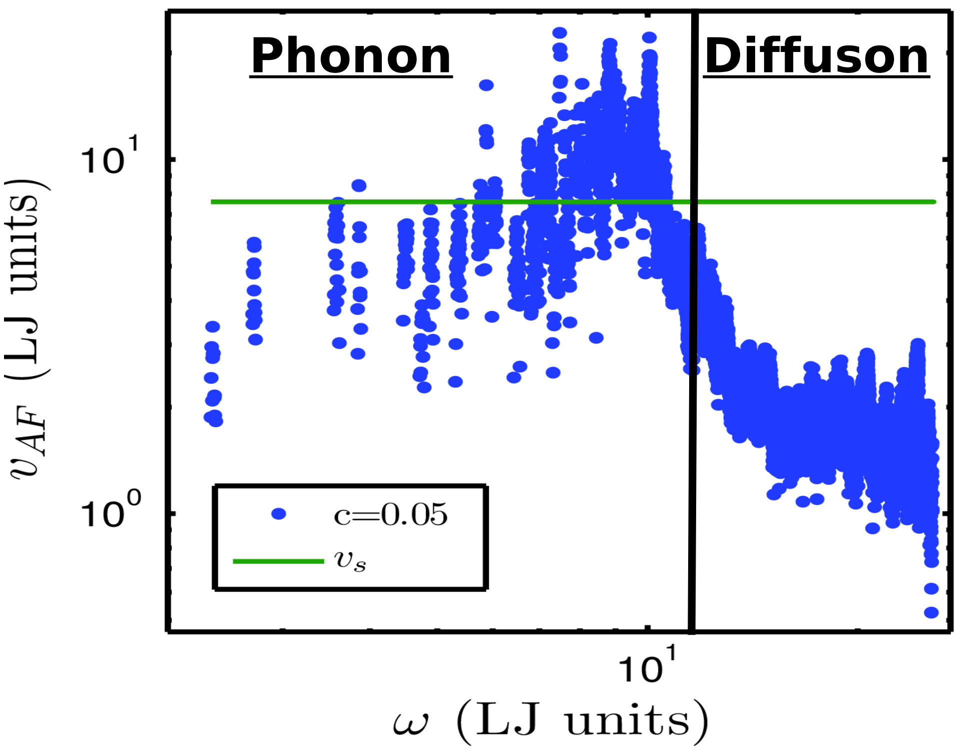
Cahill-Pohl Model:

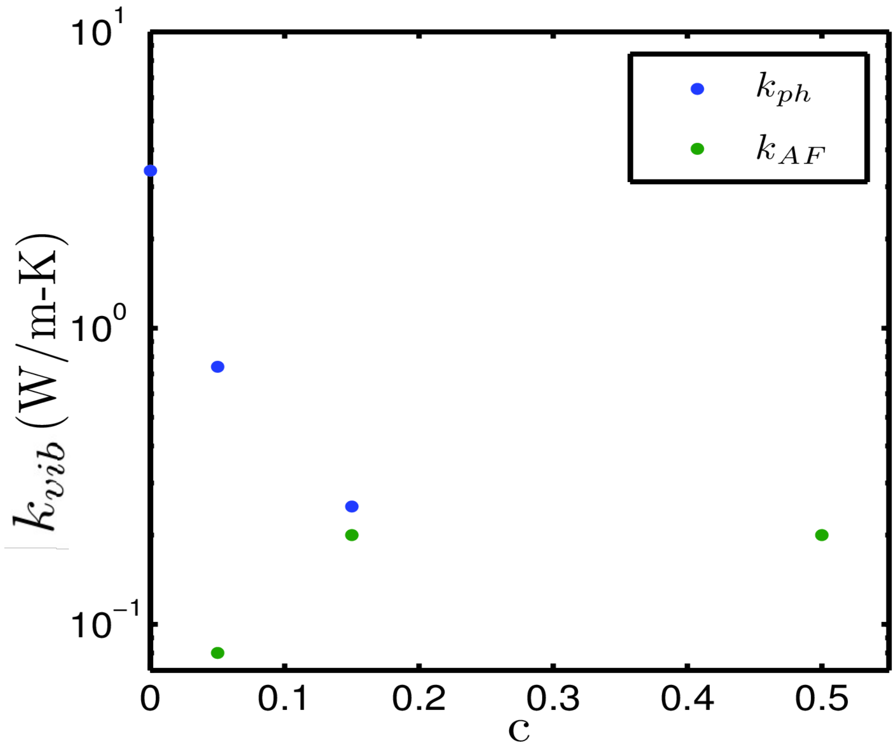
$$\tau(\omega) = 2\pi/\omega \qquad v_s \longrightarrow k_{vib}$$

Solid State Communications 70 (1989) 927-930.

Mechanical Engineering

Ordered and Disordered Vibrations





$$v_{AF}^2(\omega) = D_{AF}(\omega)/\tau(\omega)$$

v_s = sound speed

For amorphous:

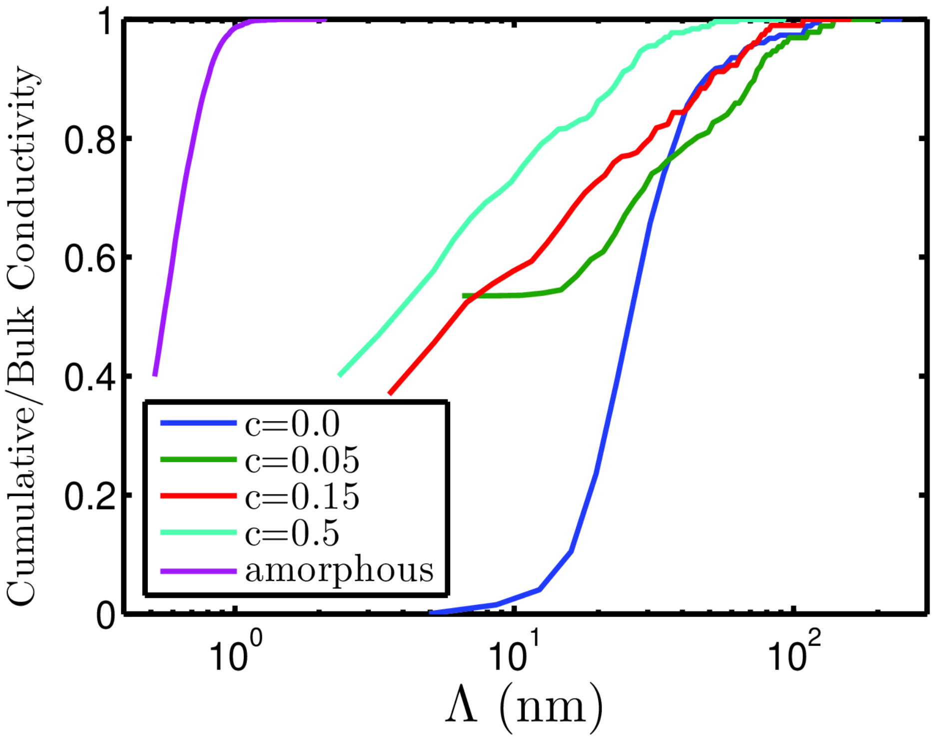
$$k_{vib} = k_{AF}$$

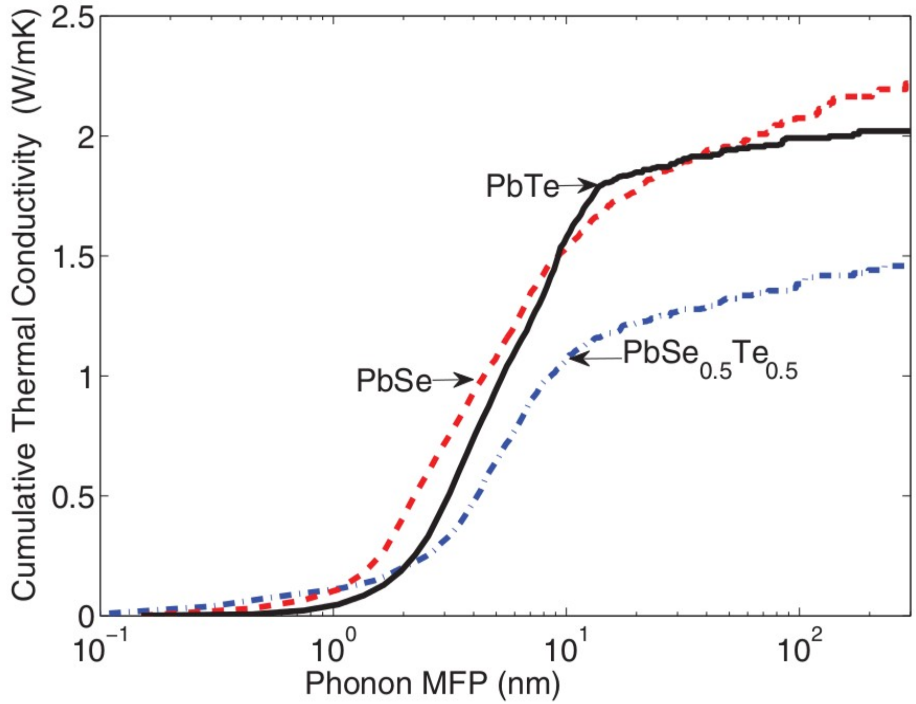
Phonon: $\Lambda = v_s \tau(\omega)$

Diffuson: $\Lambda = (D_{AF}(\omega)\tau(\omega))^{1/2}$

Mechanical Engineering

Cumulative Thermal Conductivity





PHYSICAL REVIEW B 85, 184303 (2012)

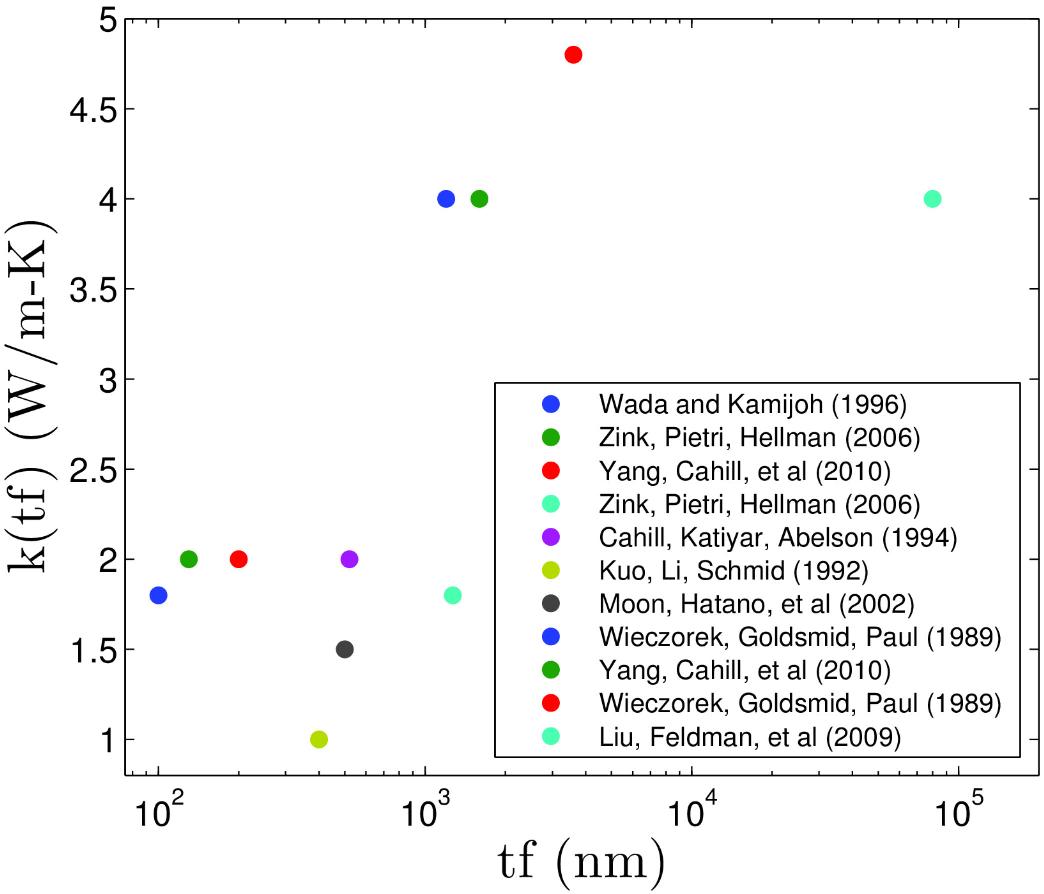
- Large c alloys and amorphous vibrations have (drastically) decreased MFP

- Boundary scattering less effective for length scales >10 nm, alloying can still be effective.

Amorphous Silicon

- a-Si thermal conductivity with varying film thickness indicates a phonon-like boundary scattering dependance.

- Ordered/Disordered analysis could measure the MFP spectrum in a-Si



$$k_{vib} = k_{AF} + k_{ph}$$

APPLIED PHYSICS LETTERS 98, 144101 2011

Mechanical Engineering

Questions

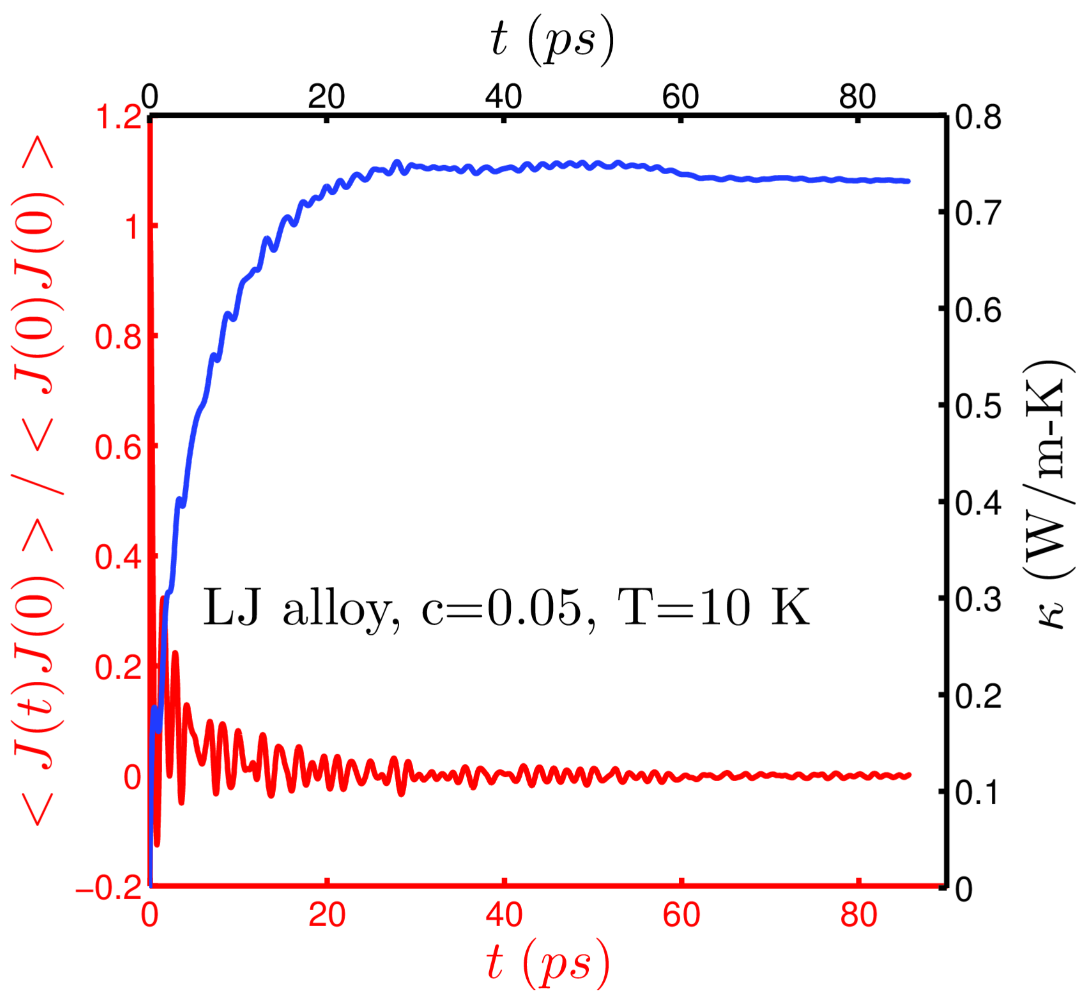
Modeling Tools

	Predicted quantities	Computational cost	Code availability
Green-Kubo (**GK**) w/ Molecular Dynamics (**MD**)	Thermal conductivity	Classical	Several
Harmonic Lattice Dynamics (**HLD**)	Vibrational frequencies, eigenvectors, group velocities, diffuson properties (Allen-Feldman (AF))	Classical/Ab-Initio	Several
Normal Mode Decomposition (**NMD**) w/ HLD and MD	Thermal conductivity, Vibrational frequencies, lifetimes	Classical	None

Mechanical Engineering

Green-Kubo

$$\kappa = \frac{V}{3k_B T^2} \int_0^\infty \langle \mathbf{J}(0) \cdot \mathbf{J}(t) \rangle \, dt$$

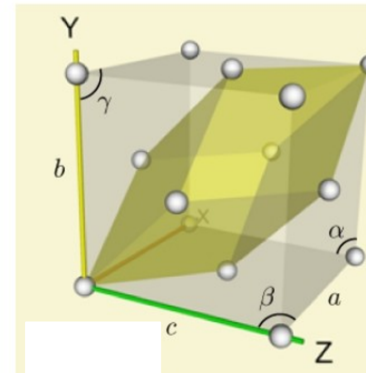


- Heat current J has all effects of MD (anharmonicity, defects, etc.)
- Heat current J has KE and PE parts.
- J is difficult to define using *ab-initio* calculations.

Mechanical Engineering

Normal Mode Decomposition (NMD)

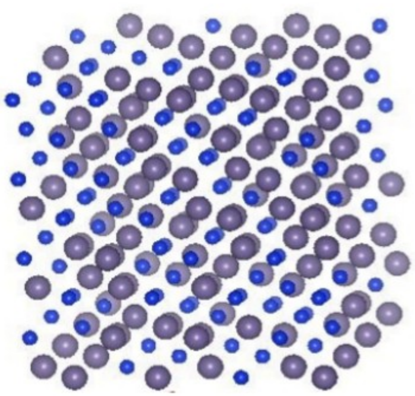
Perfect system: vibrations are phonons with an allowed wavevector $\tau\binom{\boldsymbol{\kappa}}{\nu}$

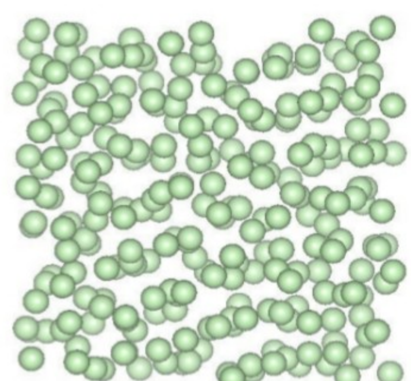


Perturbed system: vibrations are phonons with an allowed wavevector (dilute alloy). **Virtual Crystal (VC)** approximation.

Disordered system: vibrations are phonons/diffusons. Vibrations analyzed at Gamma point.

$$\tau\binom{\boldsymbol{\kappa}=0}{\omega}$$





Normal Mode Decomposition (NMD)

$$q(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix};t) = \sum_{\alpha,b,l}^{3,n,N} \sqrt{\frac{m_b}{N}} u_\alpha(\begin{smallmatrix}l\\ b\end{smallmatrix};t)\, e^*(\begin{smallmatrix}\boldsymbol{\kappa} & b\\ \nu & \alpha\end{smallmatrix}) \exp[i\boldsymbol{\kappa}\cdot\mathbf{r}_0(\begin{smallmatrix}l\\ 0\end{smallmatrix})]$$

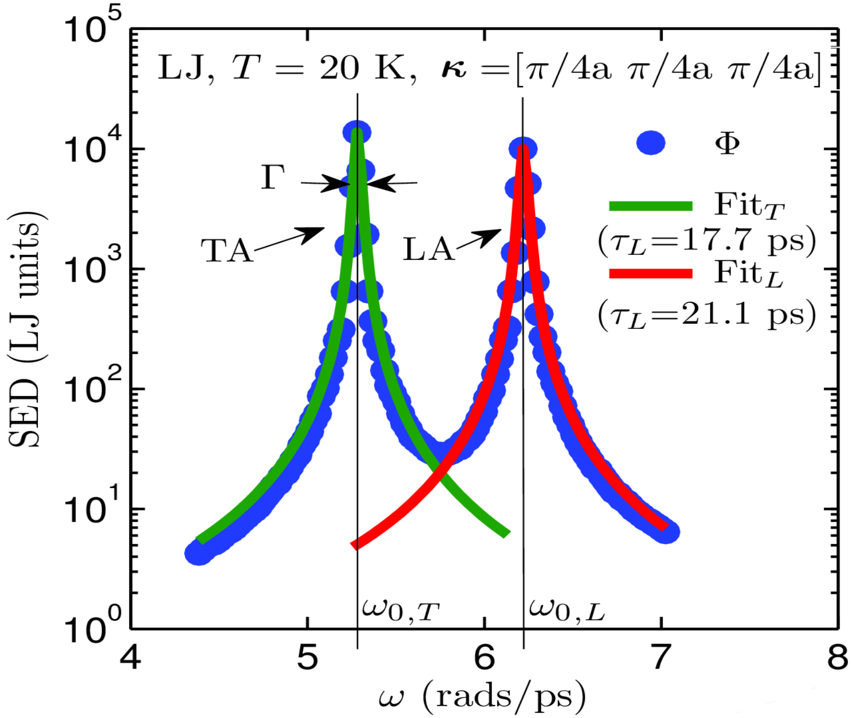
NMD: Frequency-Domain

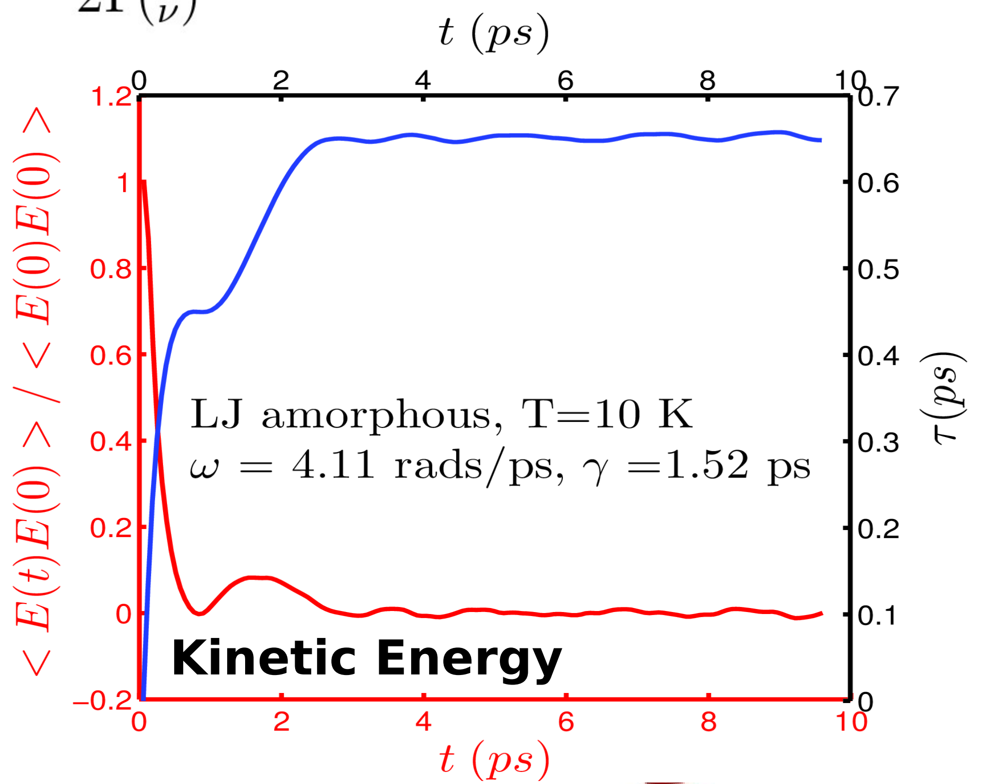
$$\Phi(\boldsymbol{\kappa},\omega) = \sum_{\nu}^{3n} C_0(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}) \frac{\Gamma(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix})/\pi}{[\omega_0(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix})-\omega]^2 + \Gamma^2(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix})}$$

NMD: Time-Domain

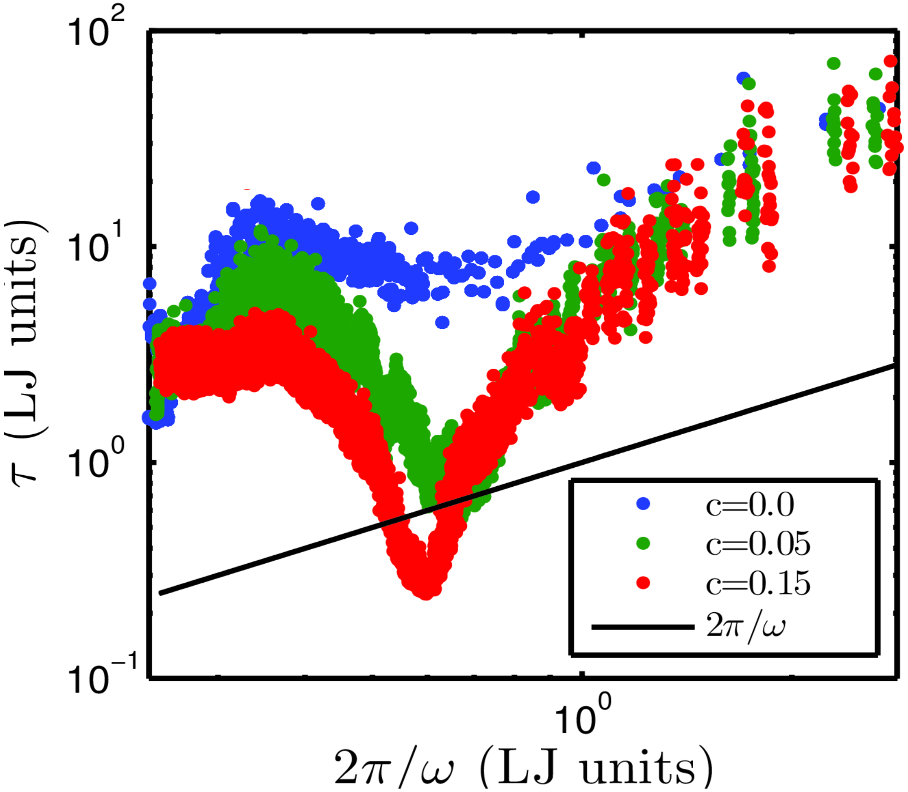
$$\exp\left[-t/\tau\begin{pmatrix}\boldsymbol{\kappa}\\ \nu\end{pmatrix}\right] = \frac{\langle E_{\boldsymbol{\kappa},\nu}(t)E_{\boldsymbol{\kappa},\nu}(0)\rangle}{\langle E_{\boldsymbol{\kappa},\nu}(0)E_{\boldsymbol{\kappa},\nu}(0)\rangle}$$

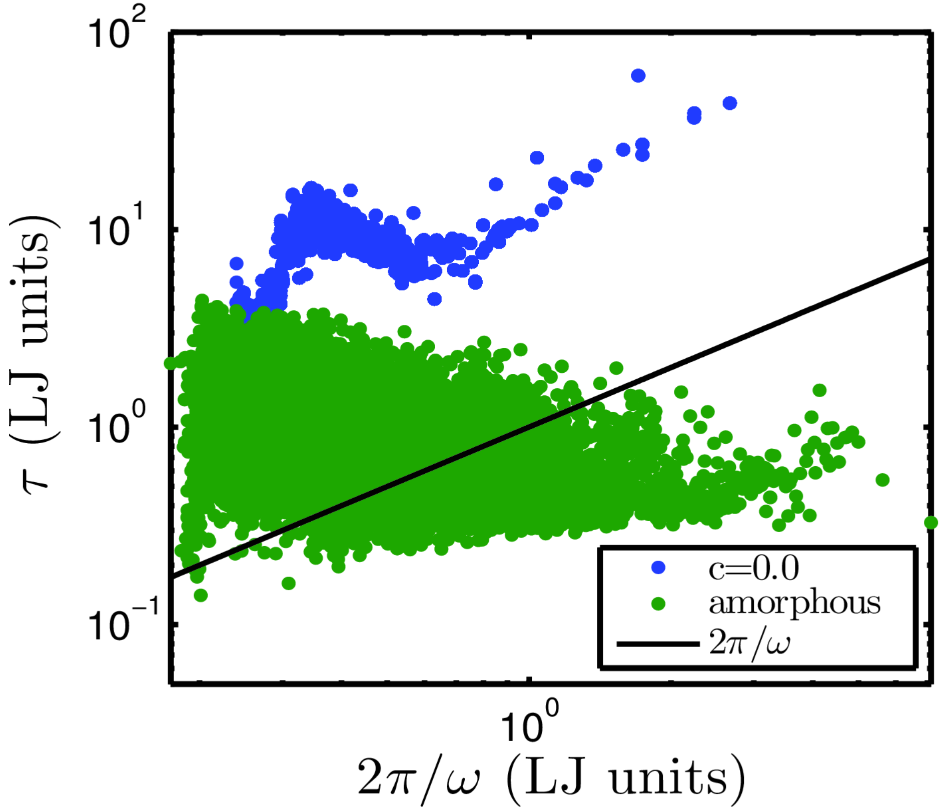
$$\tau(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}) = \frac{1}{2\Gamma(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix})}$$





Ordered and Disordered Vibrations





Ioffe-Regel Limit:

$$\tau(\omega) = 2\pi/\omega$$

PHIL. MAG. B 79, 1715-1731 (1999)

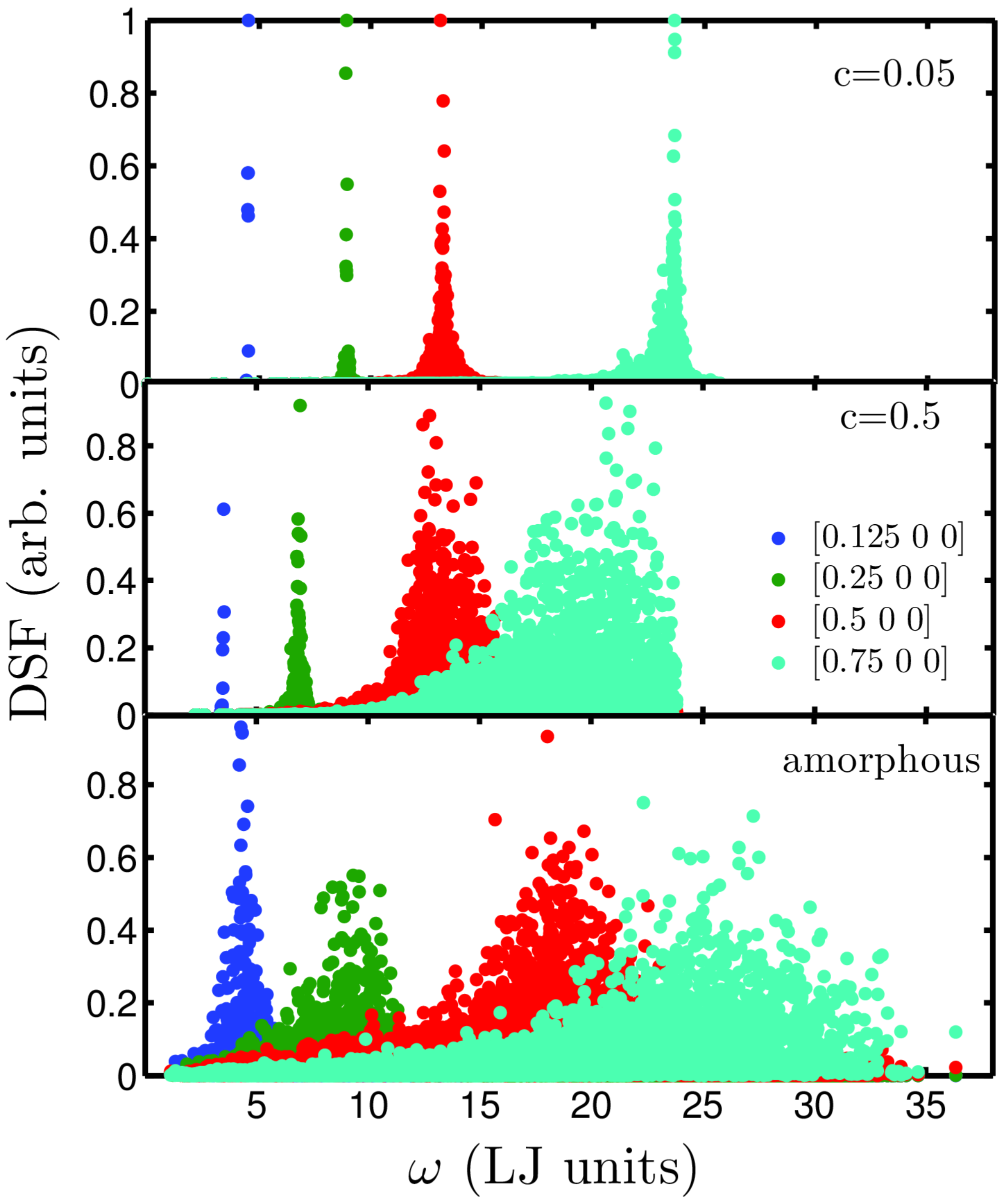
Cahill-Pohl Model:

$$\tau(\omega) = 2\pi/\omega$$

$$v_s \longrightarrow k_{vib}$$

Solid State Communications 70 (1989) 927-930.

Ordered and Disordered Vibrations



Dynamic Structure Factor:

$$S_L(\mathbf{Q},\omega)=\sum_i |A_i(\mathbf{Q})|^2\delta(\omega-\omega_i)$$

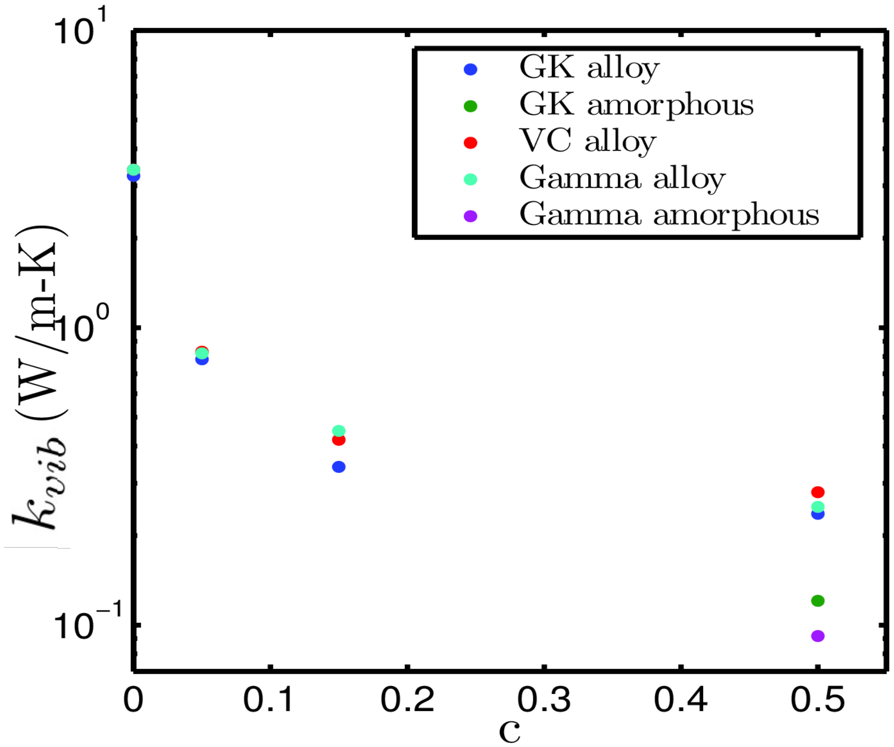
Low-frequency modes can identify a **wavelength**, not possible in general:

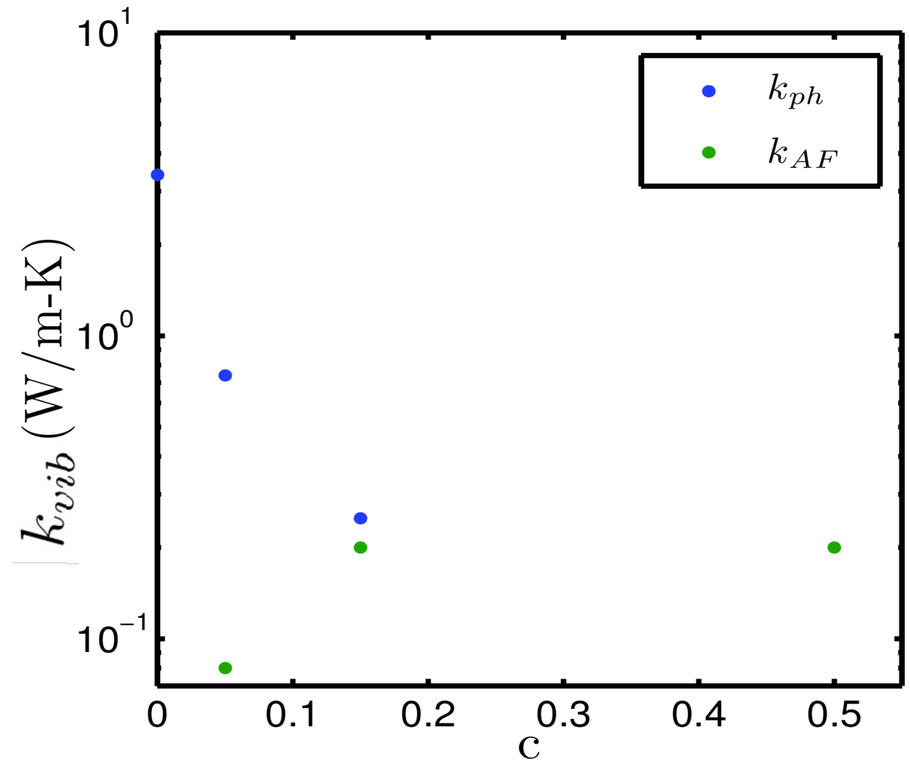
PHYS. REV. B 48, 589-601 (1993)
PHIL. MAG. B 79, 1715-1731 (1999)
PHIL. MAG. B 79, 1747-1754 (1999)

$$\lambda = ?$$

Mechanical Engineering

Predicted Thermal Conductivity





$$k_{vib} = k_{AF} + k_{ph}$$

For amorphous: $$k_{vib} = k_{AF}$$

Mechanical Engineering