

Ordered and Disordered Contributions to Lattice Thermal Conductivity

Jason Larkin

Nanoscale Transport Phenomena Laboratory
Carnegie Mellon Department of Mechanical
Engineering

<http://ntpl.me.cmu.edu/>

07/12/2012

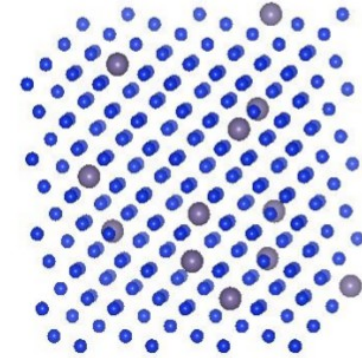


Mechanical Engineering

Thermal Transport in Disordered Materials

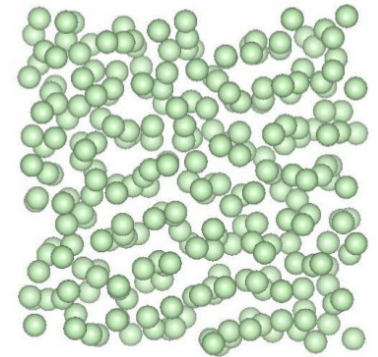
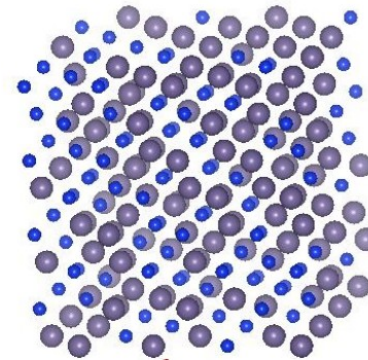
2

- **Phonon** picture valid with perturbations.



- Phonon picture only valid for very long wavelengths.

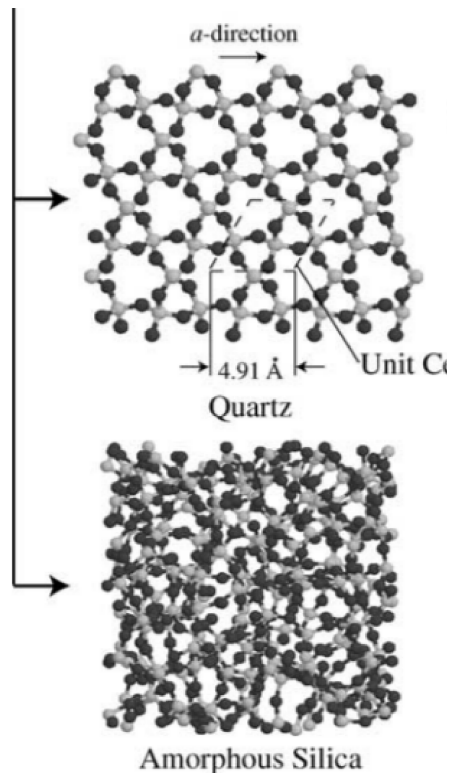
- **Localized** vibrations (diffusons) become important.



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

(disorder) (order)

Thermal Transport in Disordered Materials: Applications



+Disorder

Crystal	σ	K
	HI	HI
LUC		
	HI	LO
Glass		
	LO	LO

$$ZT = \frac{S^2 \sigma T}{k_{thermal}}$$



Theoretical Models

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

Phonons (ordered): Interacting Phonon Gas

$$k_{ph} = \sum_{\kappa} \sum_{\nu} c_{ph}(\kappa_{\nu}) v_{g,\mathbf{n}}^2(\kappa_{\nu}) \tau(\kappa_{\nu}) \quad \Lambda(\kappa_{\nu}) = |\mathbf{v}_g| \tau(\kappa_{\nu})$$

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

Diffusons (disordered): Allen-Feldman Theory

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

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

$$C(\omega_i) = k_B \quad c_{ph}(\kappa_{\nu}) = k_B$$

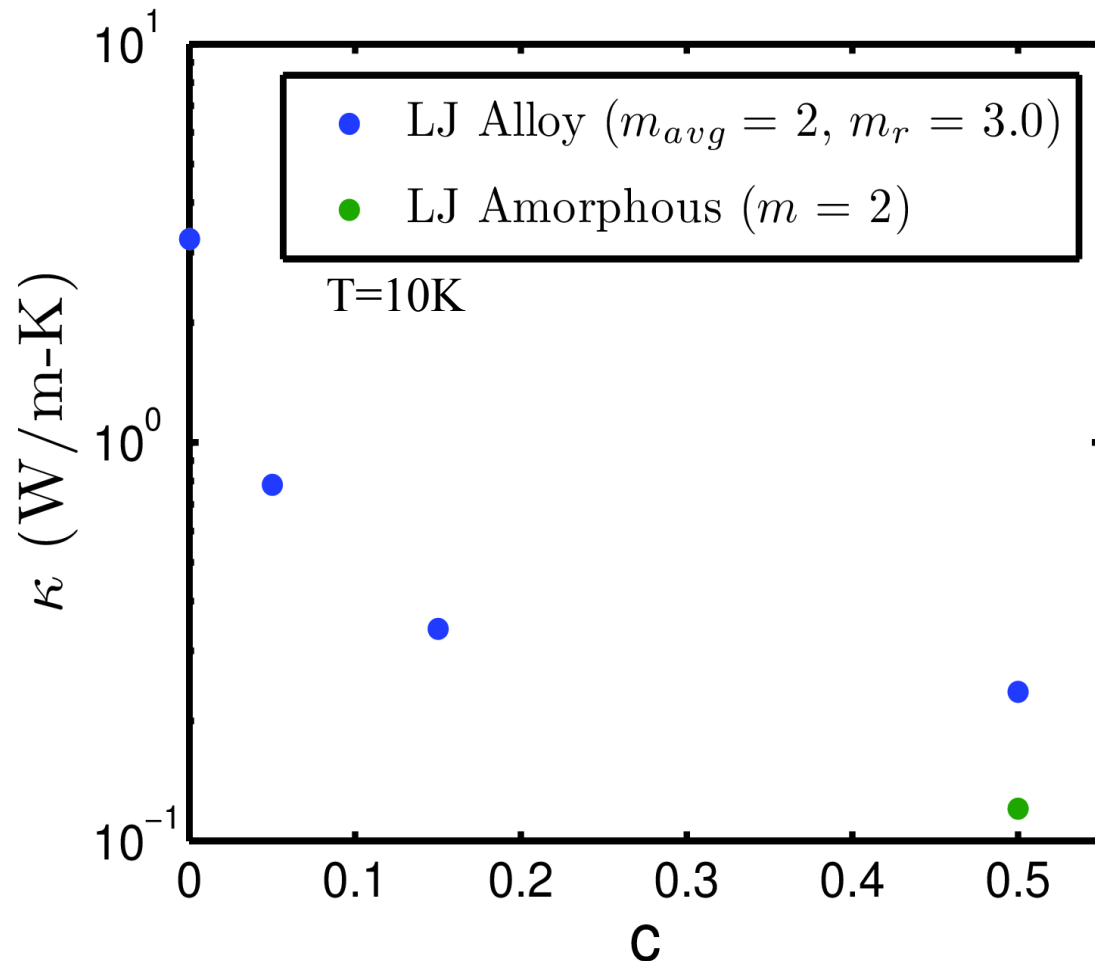
Modeling Tools

	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, diffusion properties (Allen-Feldman (AF))	Classical/Ab-Initio	Several
Normal Mode Decomposition (NMD) w/ HLD and MD	Thermal conductivity, Vibrational frequencies, lifetimes	Classical	None

Thermal Conductivity: System-Level

Green-Kubo

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



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

Normal Mode Decomposition (NMD)

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

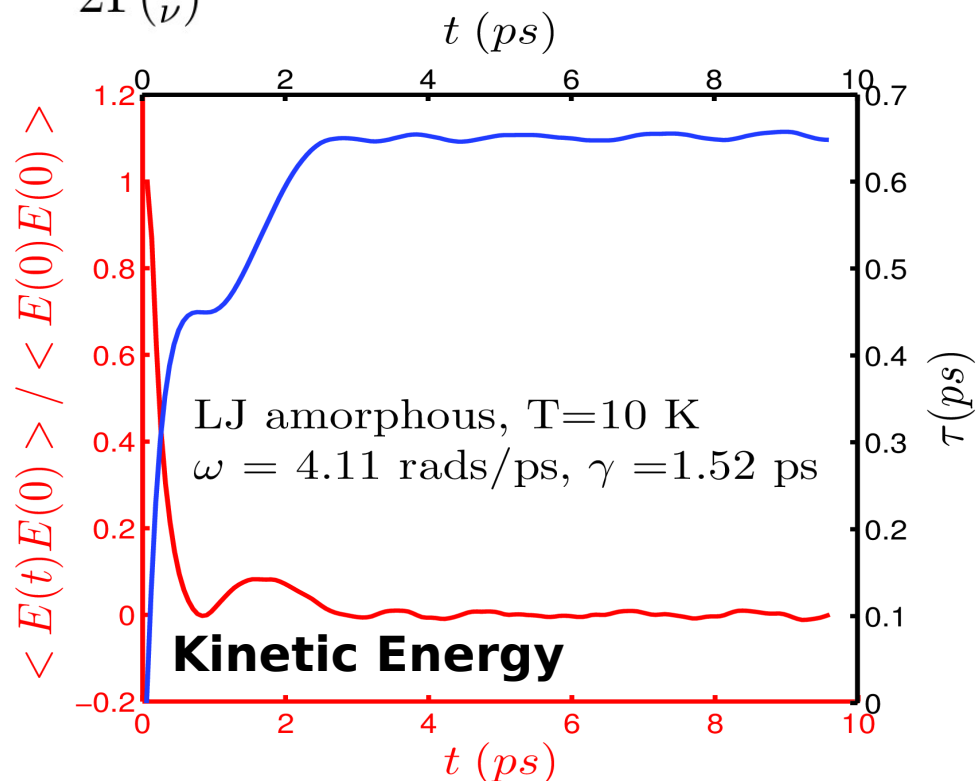
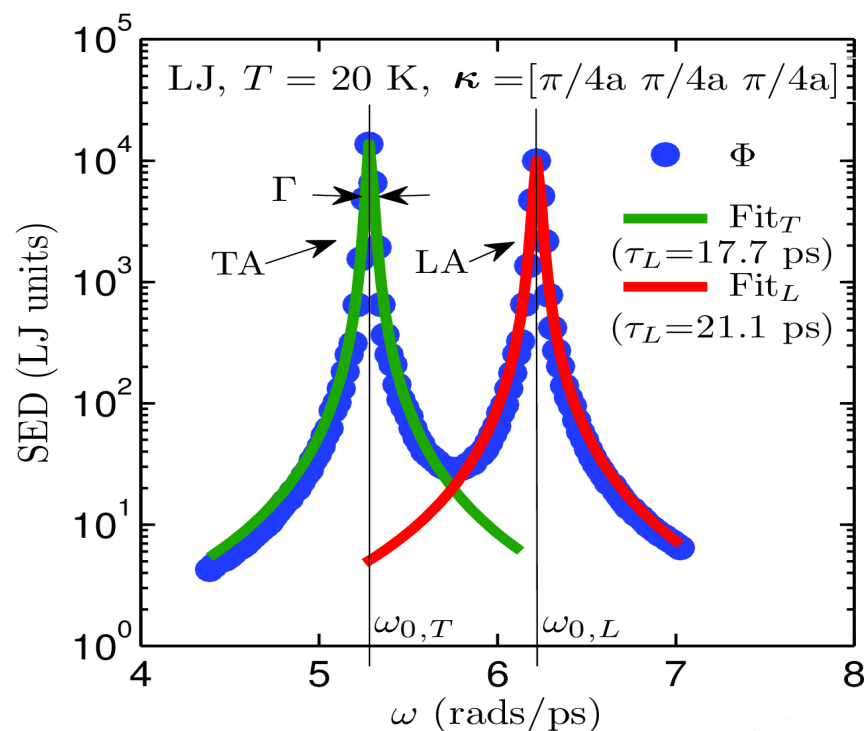
NMD: Frequency-Domain

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

NMD: Time-Domain

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

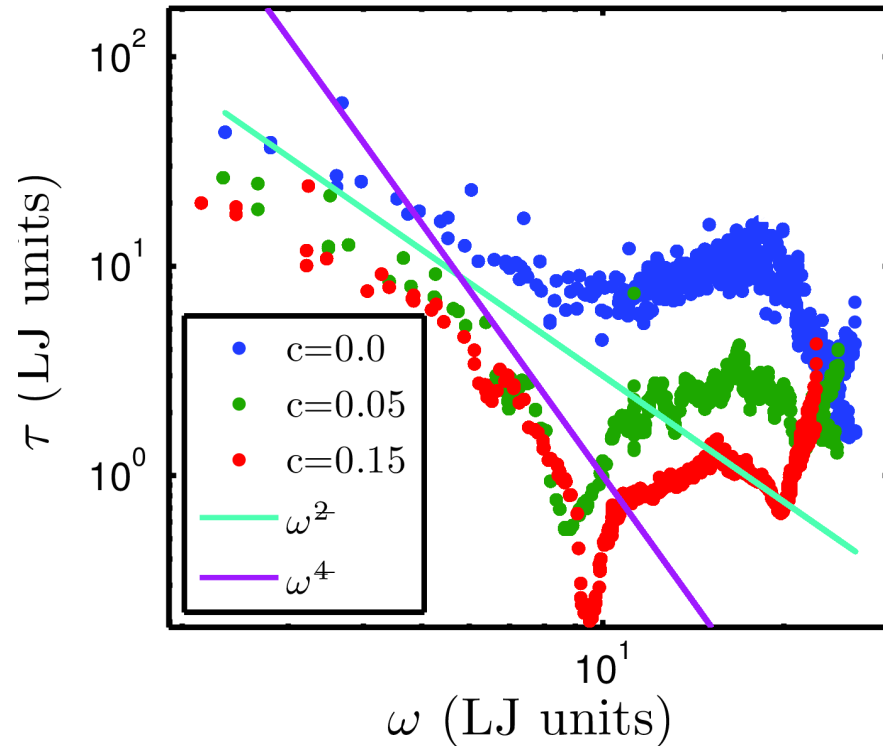
$$\tau(\boldsymbol{\kappa}_{\nu}) = \frac{1}{2\Gamma(\boldsymbol{\kappa}_{\nu})}$$



Normal Mode Decomposition (NMD)

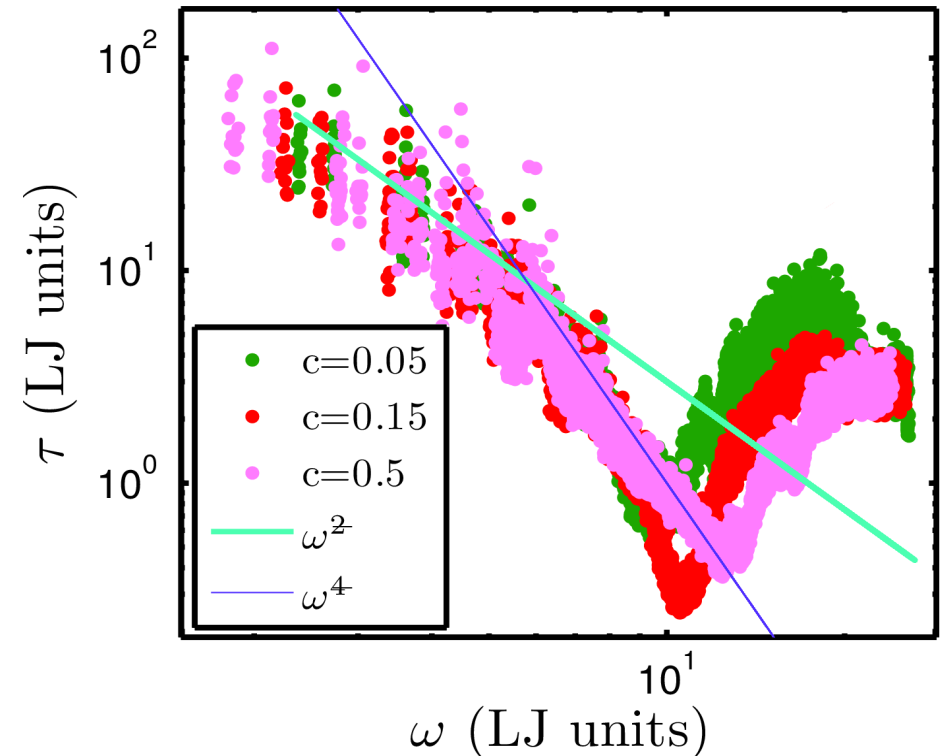
8

Virtual Crystal



$$\tau(\kappa_\nu)$$

Gamma point

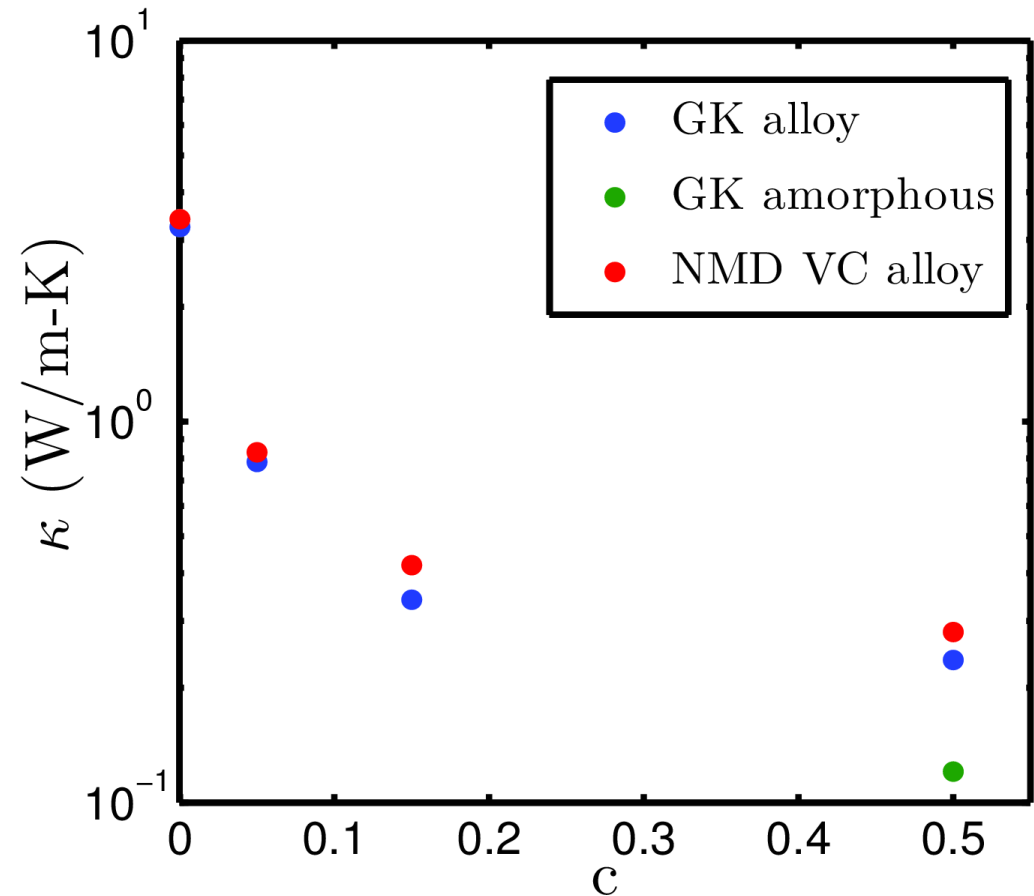


$$\tau(\kappa_\omega = 0)$$

Thermal Conductivity: System- and Carrier-Level

Virtual Crystal approximation and phonon scaling relations work well!

Anharmonic Lattice Dynamics + Defect scaling =



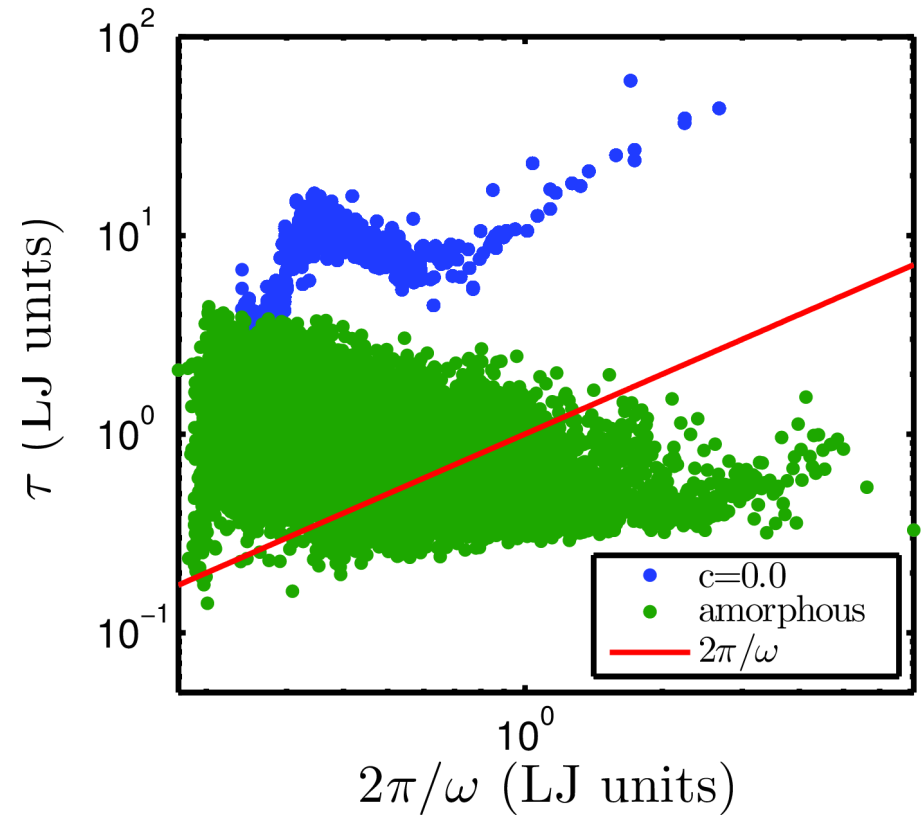
PHYSICAL REVIEW B 85, 184303 (2012)

PRL 106, 045901 (2011)

Ordered and Disordered Vibrations

Ioffe-Regel Limit:

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



Cahill-Pohl Model:

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

Solid State Communications 70 (1989) 927-930.

Ordered and Disordered Vibrations

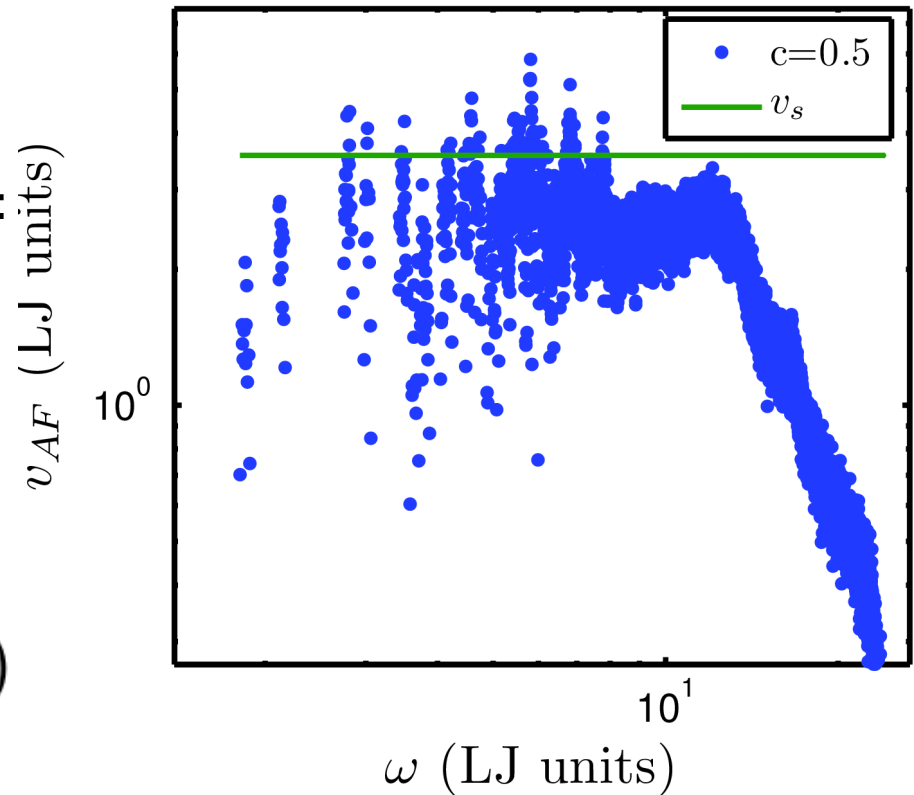
Dynamic Structure Factor:
Low-frequency modes can identify a
wavelength, not possible in general:

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

PHIL. MAG. B 79, 1747-1754 (1999)

$$\Lambda = ?$$

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



If: $v_{AF}(\omega) > v_s$ $\Lambda = v_s \tau(\omega)$

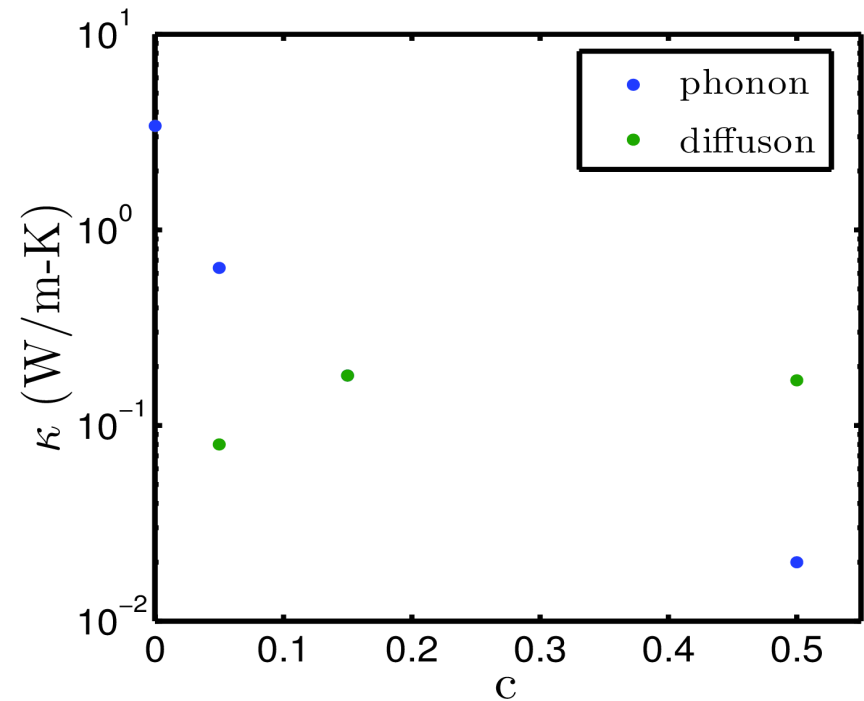
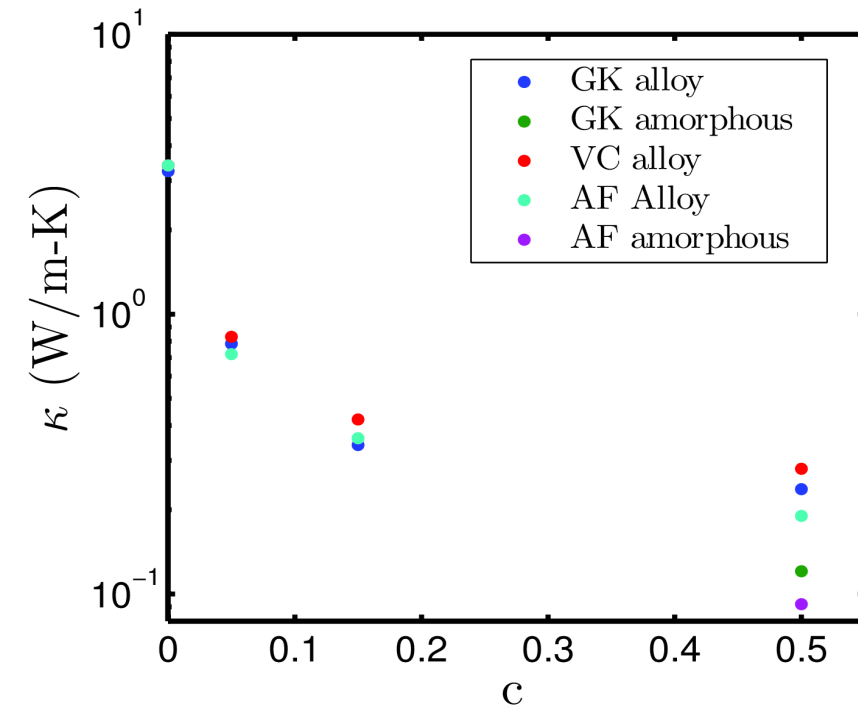
Phonon

Else: $v_{AF}(\omega) < v_s$ $\Lambda = (D_{AF}(\omega) \tau(\omega))^{1/2}$ **Diffuson**



Predicted Thermal Conductivity

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

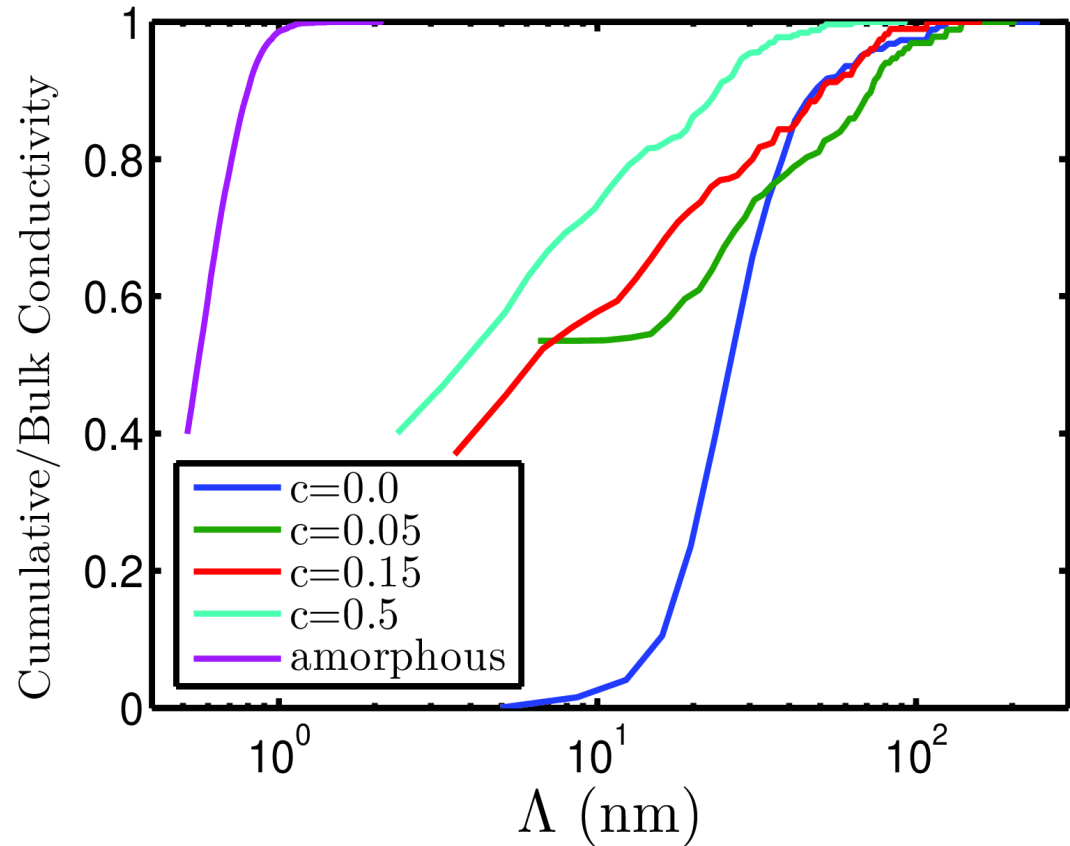


Cumulative Thermal Conductivity

Boundary Scattering:
Cumulative Thermal
Conductivity

Large c alloys and
amorphous vibrations
have decreased MFP

Boundary scattering less
effective for >10 nm

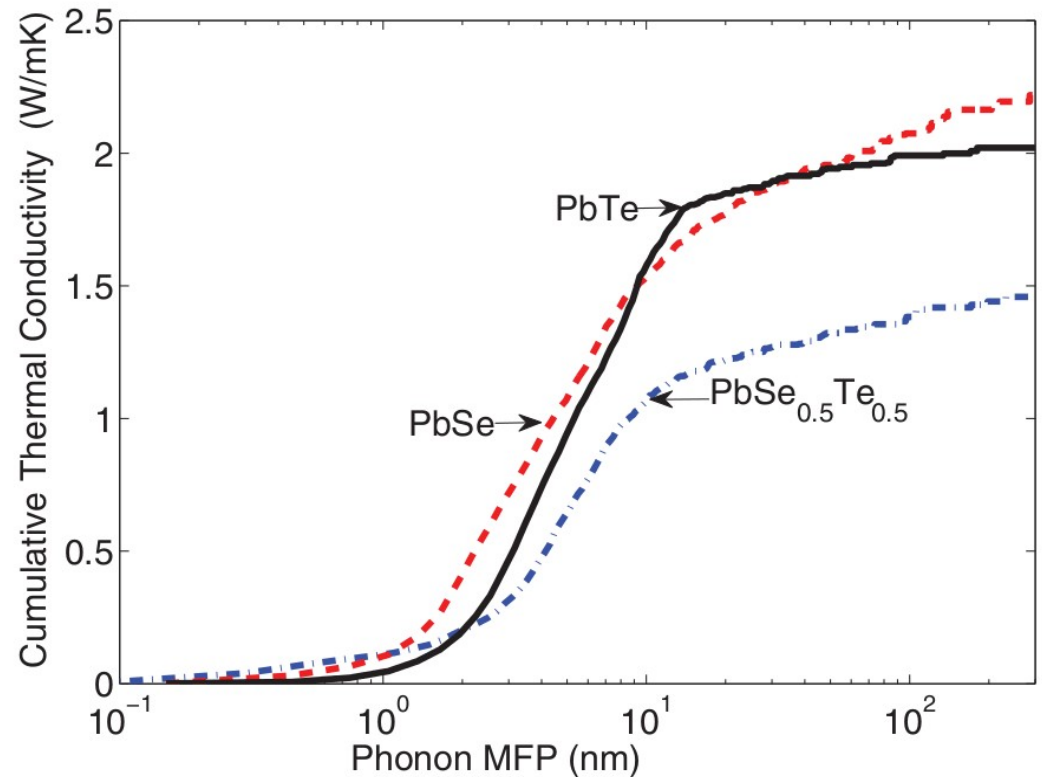


Bulk Thermoelectrics

For Bulk thermoelectrics, systems are “soft” (small group velocities, small lifetimes)

MFPs are typically < 100 nm, boundary scattering not effective

Alloying can still be effective



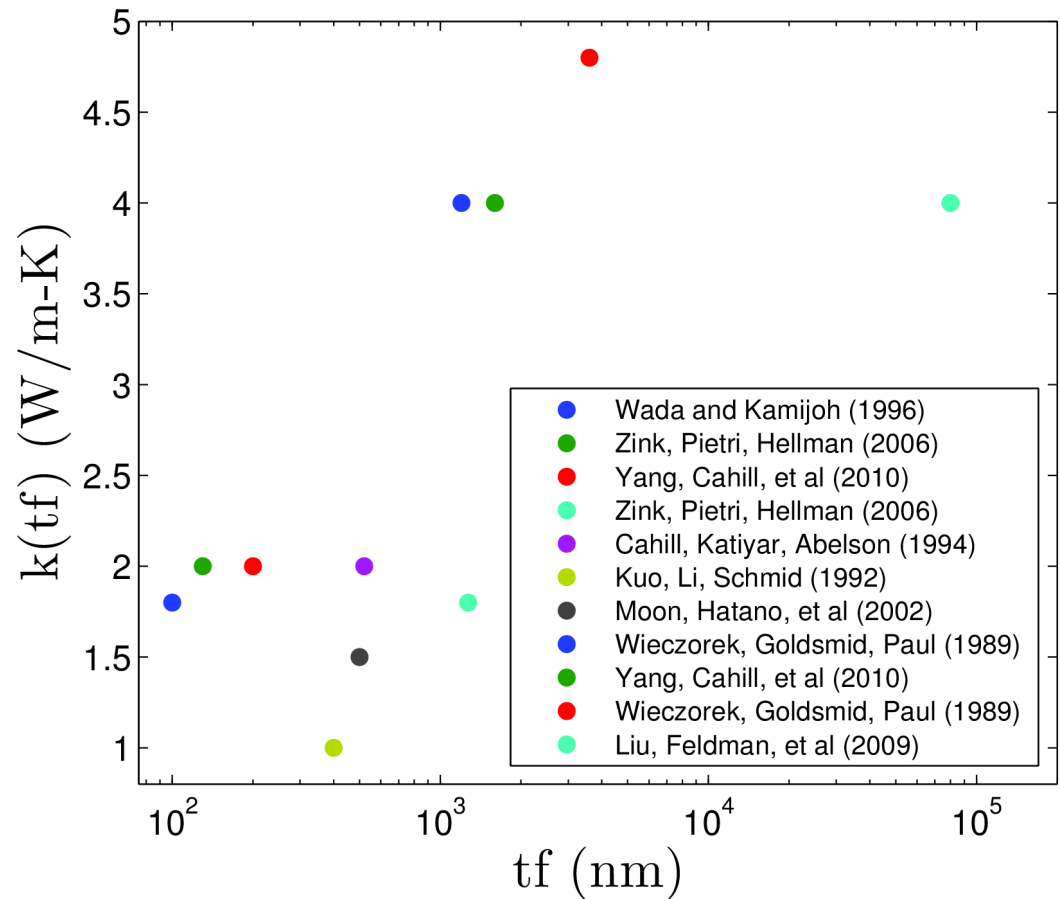
PHYSICAL REVIEW B 85, 184303 (2012)

Amorphous Silicon

In a-Si, thermal conductivity displays clear film thickness dependence

Indicates a phonon-like boundary scattering dependence

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



APPLIED PHYSICS LETTERS 98, 144101'2011

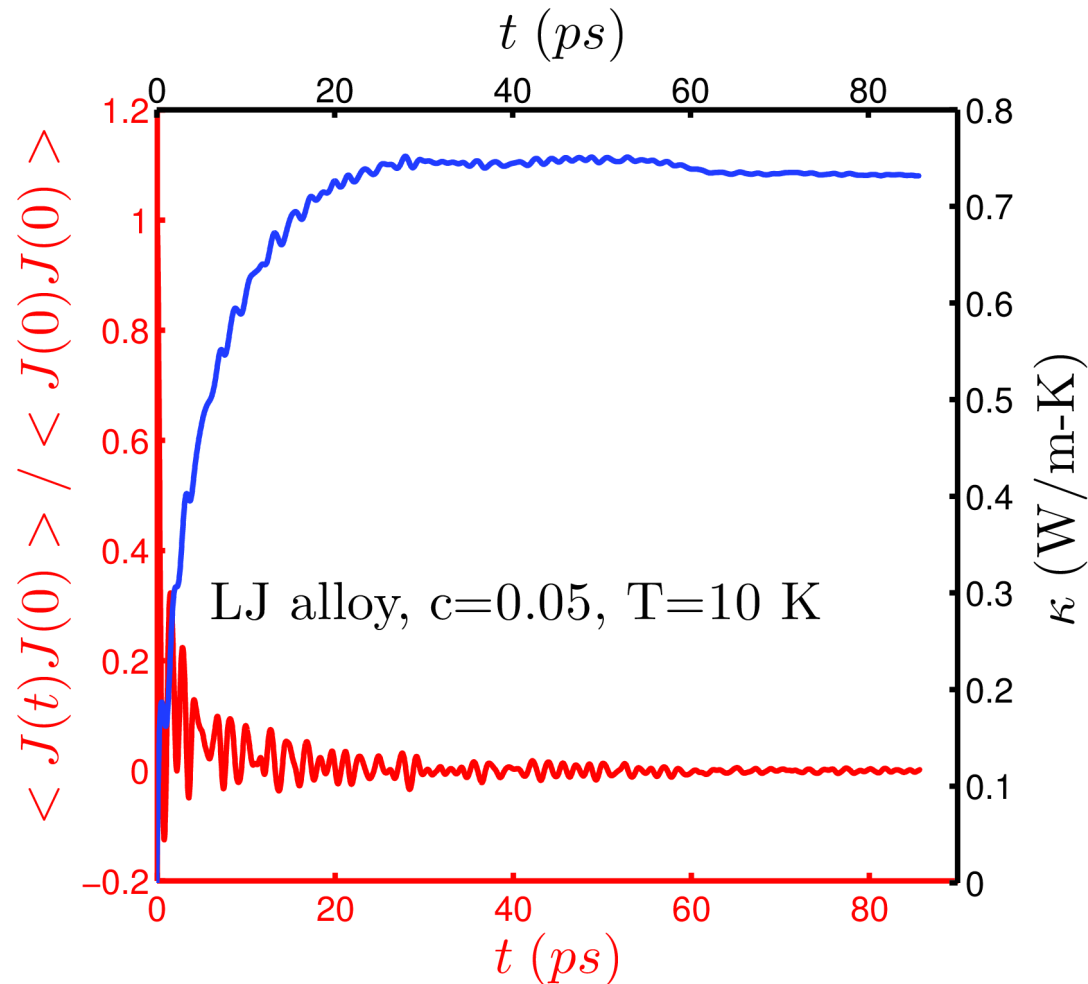
Questions

16



Green-Kubo

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



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

Normal Mode Decomposition (NMD)

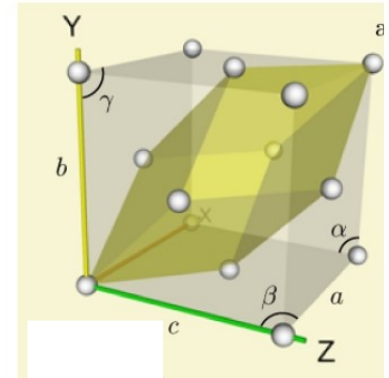
34

Perfect system: vibrations are phonons with an allowed wavevector

$$\tau\left(\begin{matrix} \kappa \\ \nu \end{matrix}\right)$$

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.

$$\tau\left(\begin{matrix} \kappa \\ \omega \end{matrix} = 0\right)$$

