Ordered and Disordered Contributions to Lattice Thermal Conductivity

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07/12/2012

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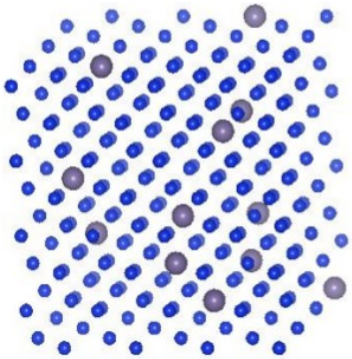
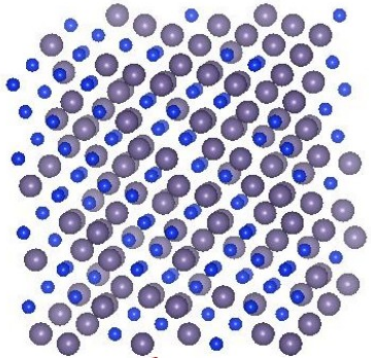
Thermal Transport in Disordered Materials

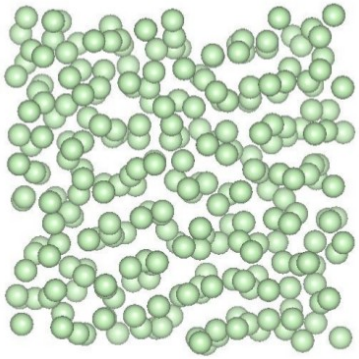
- **Phonon** picture valid with perturbations.



- Phonon picture only valid for very long wavelengths.

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

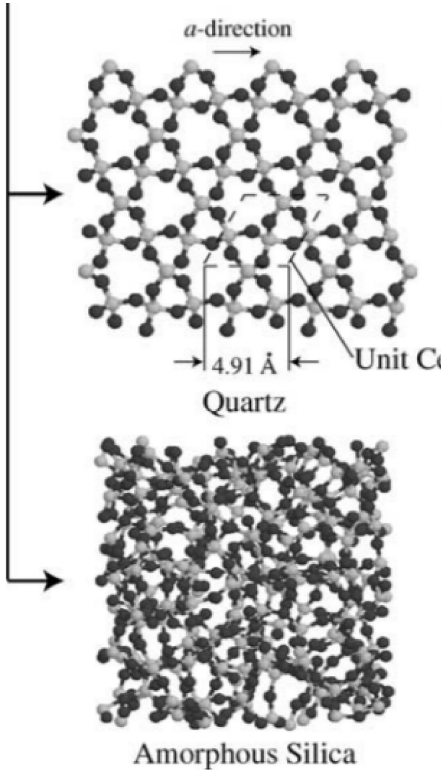


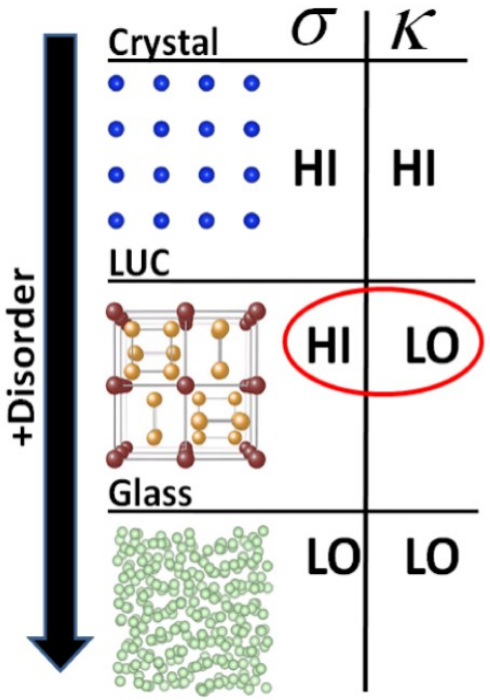


$$k_{vib} = k_{AF} + k_{ph}$$

(disorder) (order)

Thermal Transport in Disordered Materials: Applications





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

Theoretical Models

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

Phonons (ordered): Interacting Phonon Gas

$$k_{ph} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} c_{ph}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right) v^2_{g,\mathbf{n}}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right) \tau\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right) \quad \Lambda\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right) = |\boldsymbol{v}_g|\tau\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right)$$

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

Diffusons (disordered): Allen-Feldman Theory

$$k_{AF} = \sum_{i} C(\omega_i) D_{AF}(\omega_i) \qquad \Lambda = \ ?$$

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

$$C(\omega_i) = k_B \qquad c_{ph}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right) = k_B$$

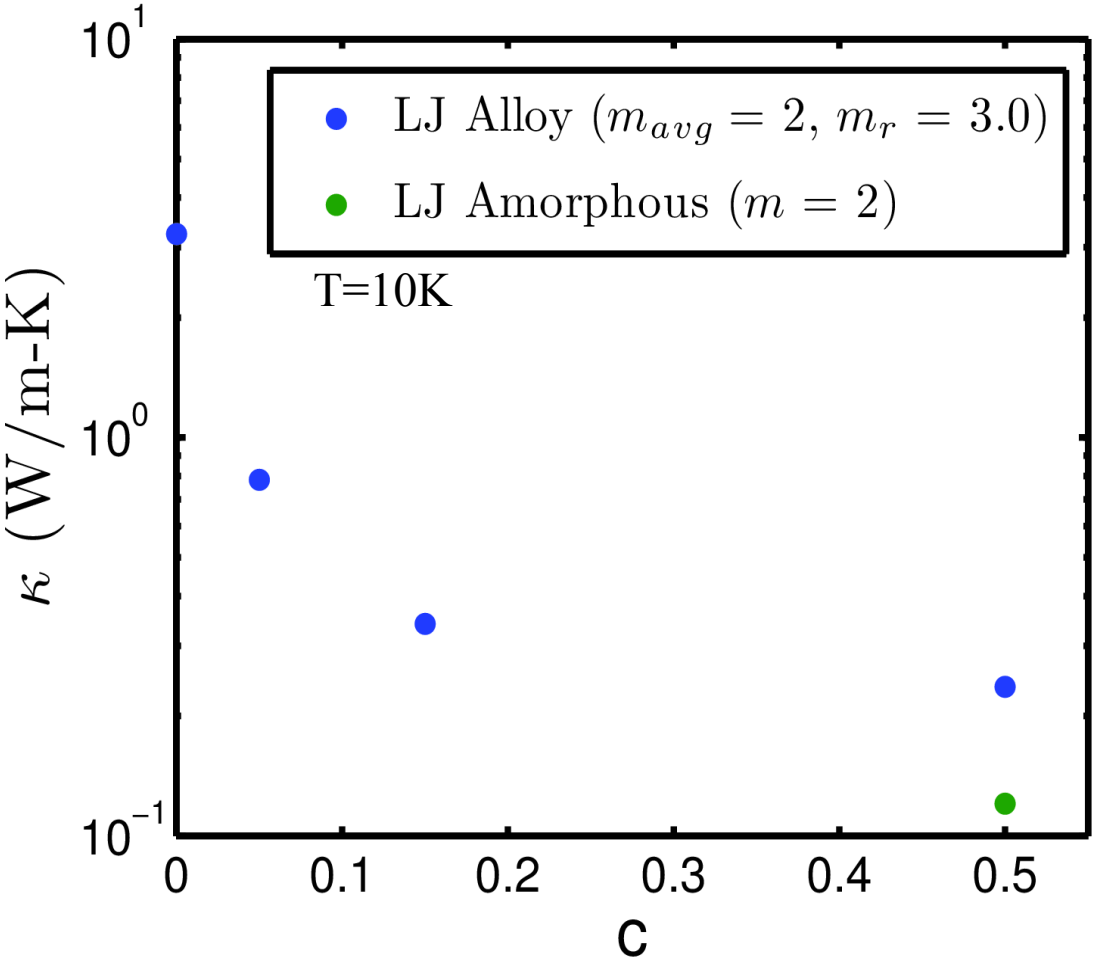
Modeling Tools

	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, diffuson 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 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.

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Normal Mode Decomposition (NMD)

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

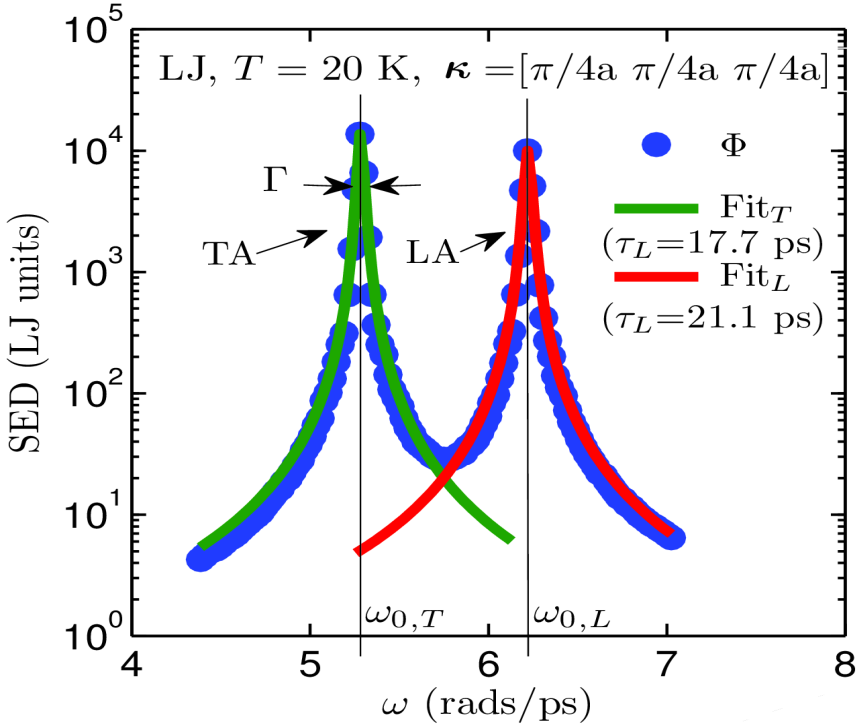
NMD: Frequency-Domain

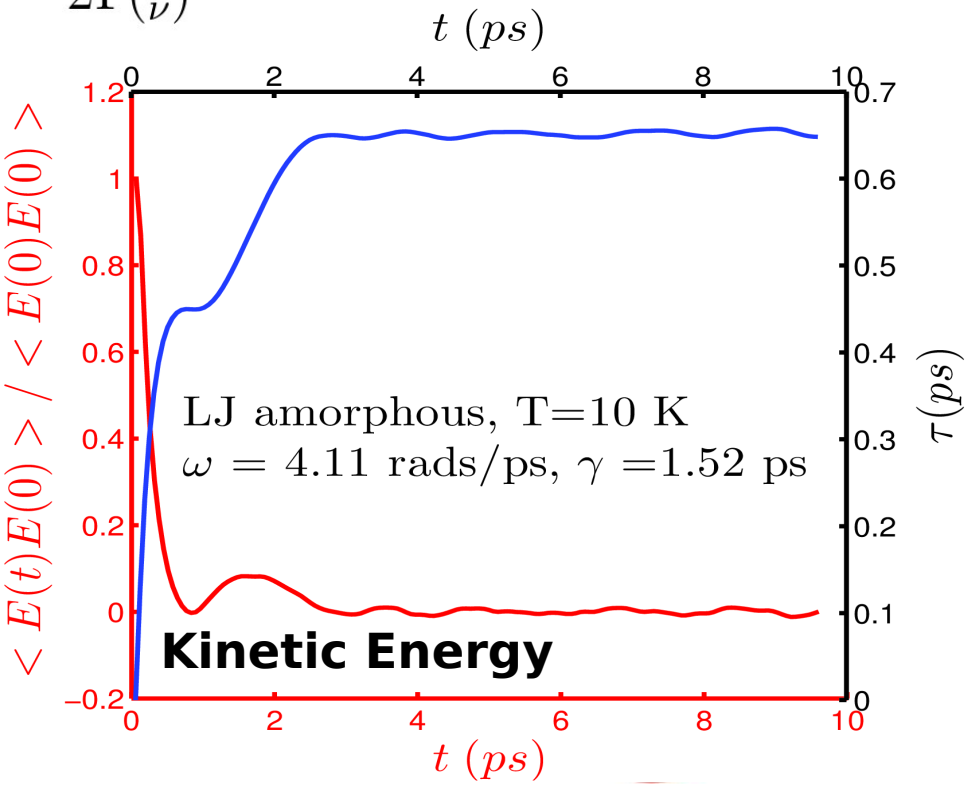
$$\Phi(\boldsymbol{\kappa},\omega) = \sum_{\nu}^{3n} C_0(\kappa;\nu)\, \frac{\Gamma(\kappa;\nu)/\pi}{[\omega_0(\kappa;\nu)-\omega]^2 + \Gamma^2(\kappa;\nu)}$$

NMD: Time-Domain

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

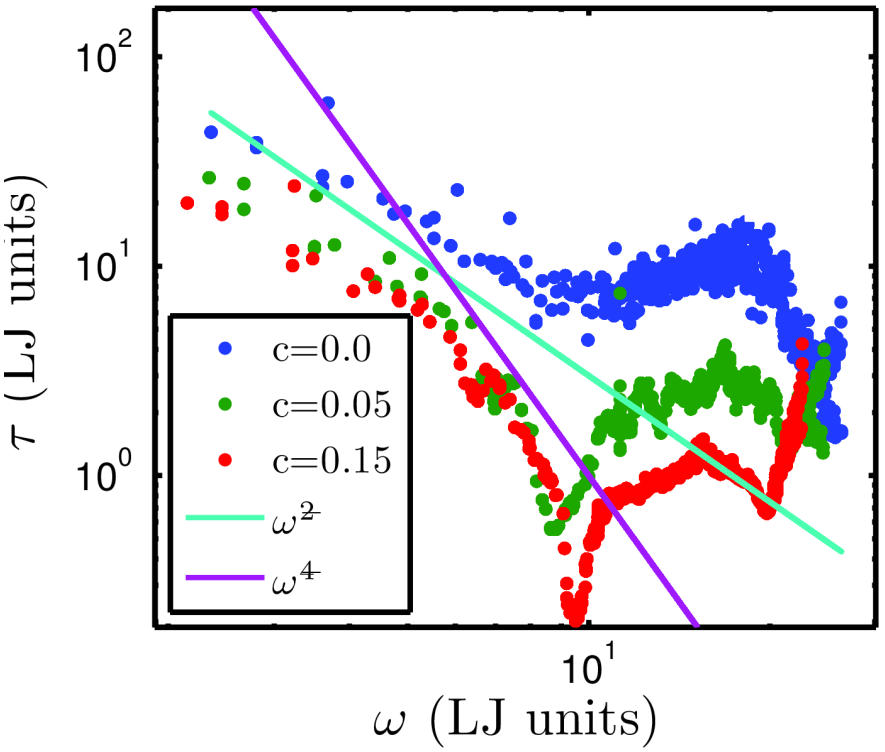
$$\tau(\kappa;\nu) = \frac{1}{2\Gamma(\kappa;\nu)}$$





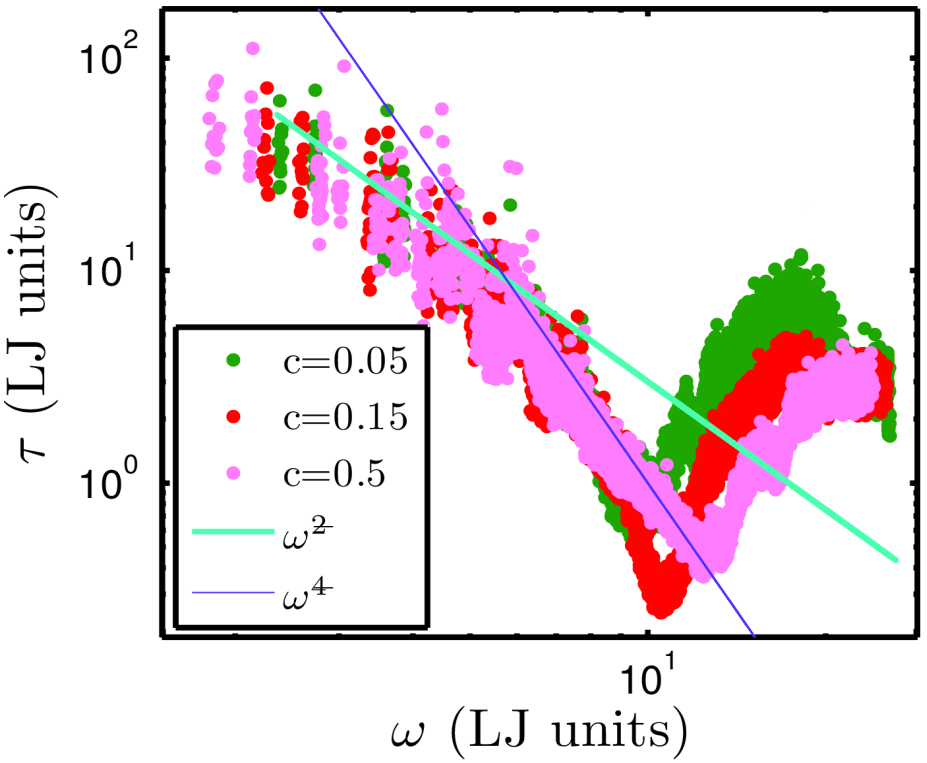
Normal Mode Decomposition (NMD)

Virtual Crystal



$$\tau\begin{pmatrix}\kappa\\ \nu\end{pmatrix}$$

Gamma point



$$\tau\begin{pmatrix}\kappa = 0\\ \omega\end{pmatrix}$$

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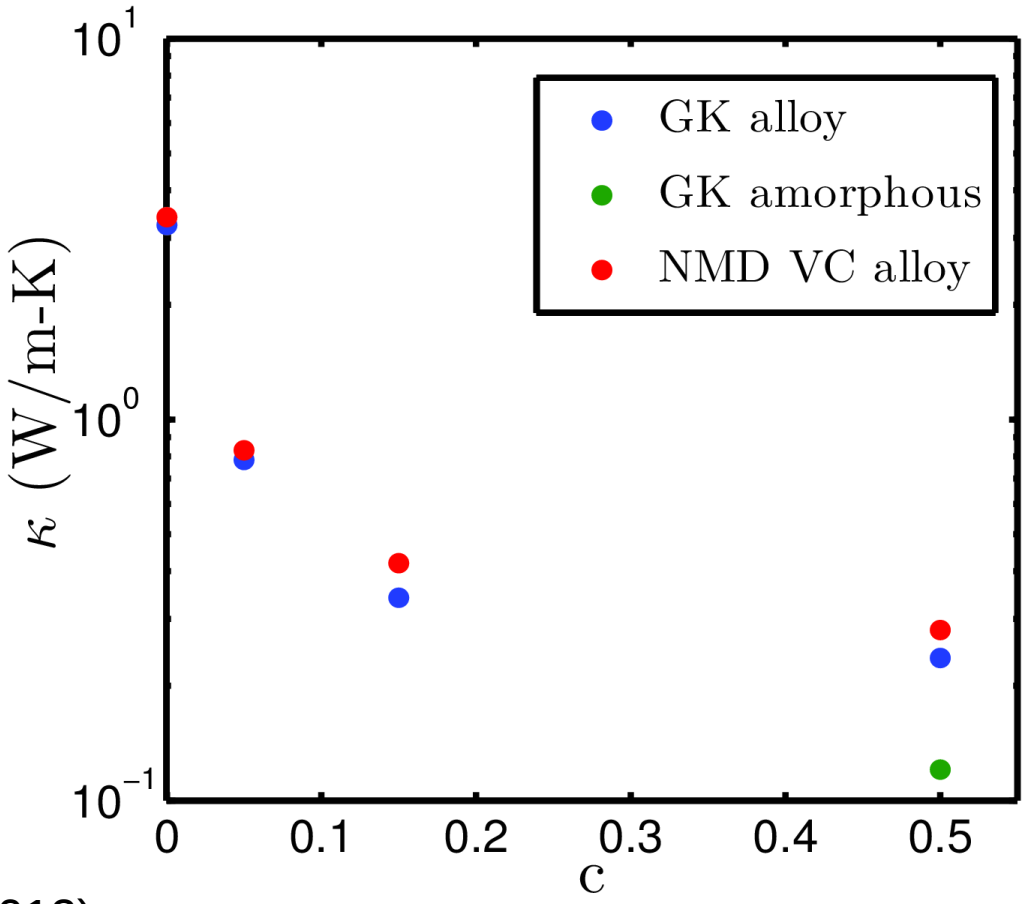
Thermal Conductivity: System- and Carrier-Level

Virtual Crystal approximation and phonon scaling relations work well!

Anharmonic Lattice Dynamics + Defect scaling =



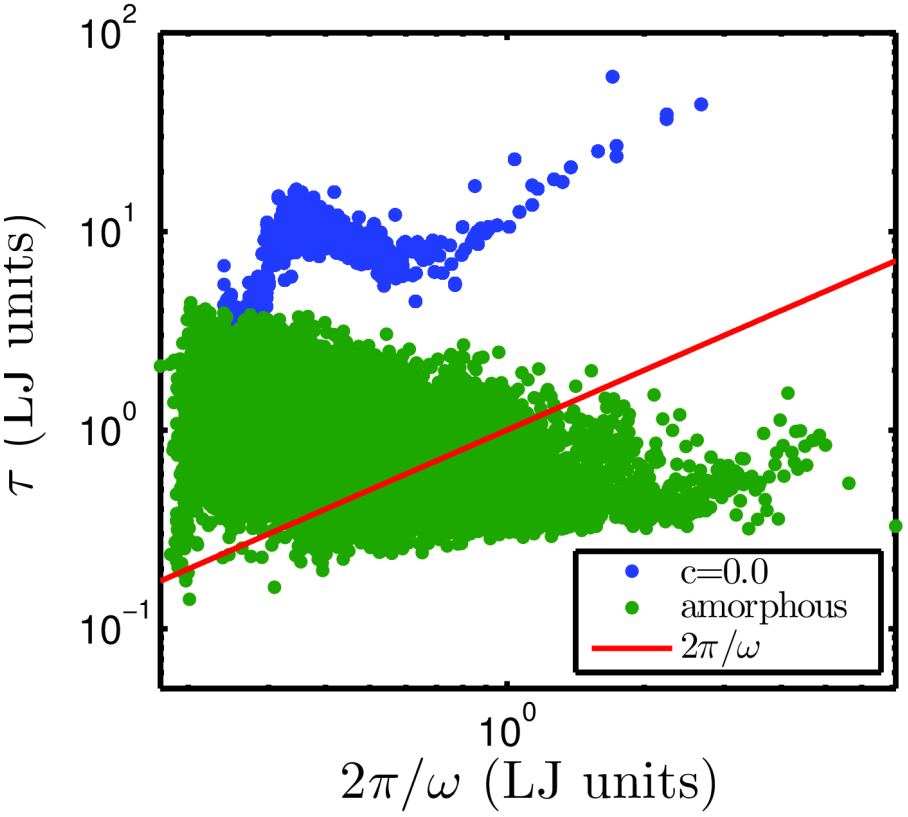
PHYSICAL REVIEW B 85, 184303 (2012)

PRL 106, 045901 (2011)

Ordered and Disordered Vibrations

Ioffe-Regel Limit:

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



Cahill-Pohl Model:

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

Solid State Communications 70 (1989) 927-930.

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Ordered and Disordered Vibrations

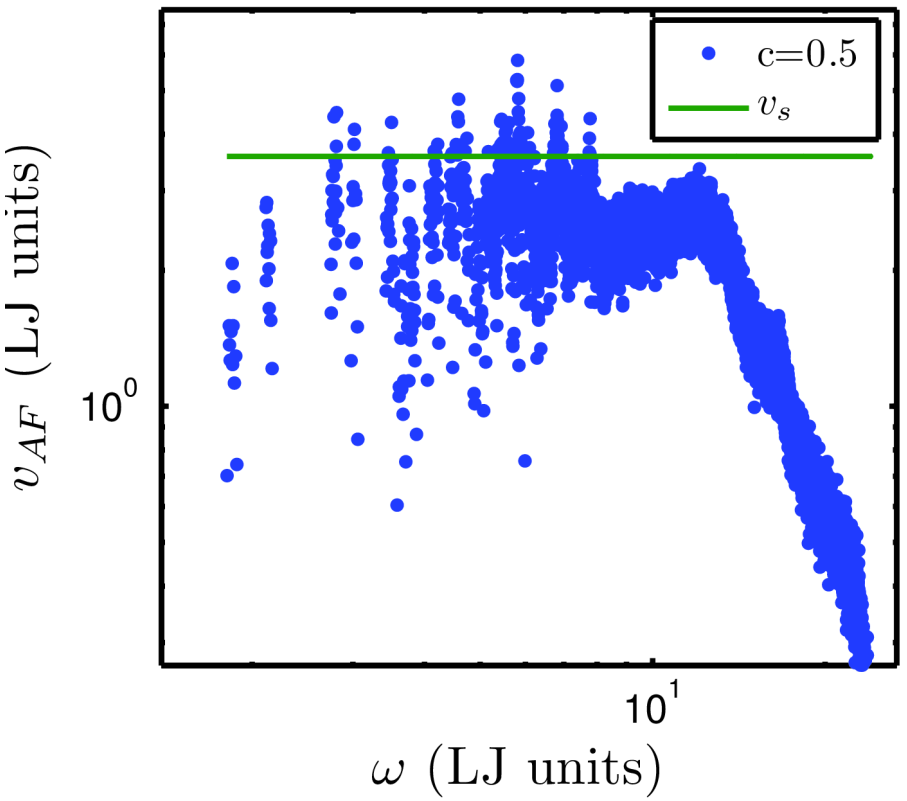
Dynamic Structure Factor:
Low-frequency modes can identify a **wavelength**, not possible in general:

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

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

$$\Lambda = \ ?$$

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

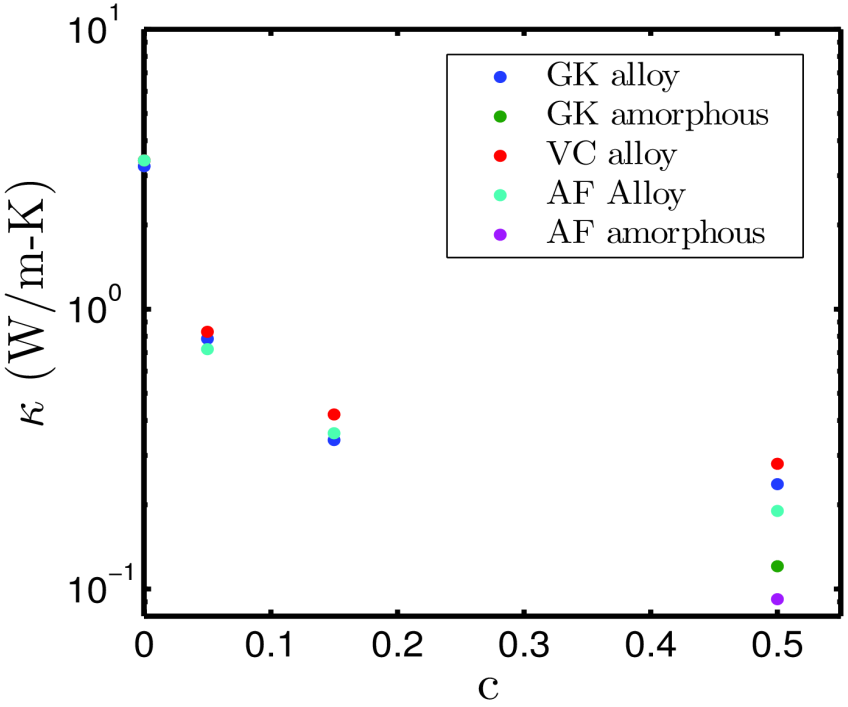


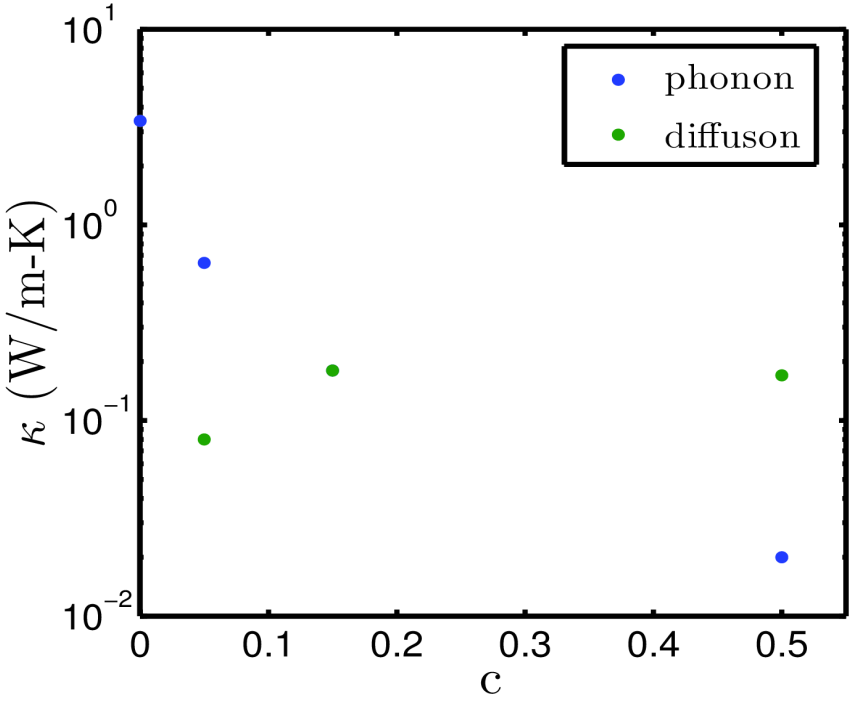
If: $v_{AF}(\omega) > v_s$ $\Lambda = v_s \tau(\omega)$ **Phonon**

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

Predicted Thermal Conductivity

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



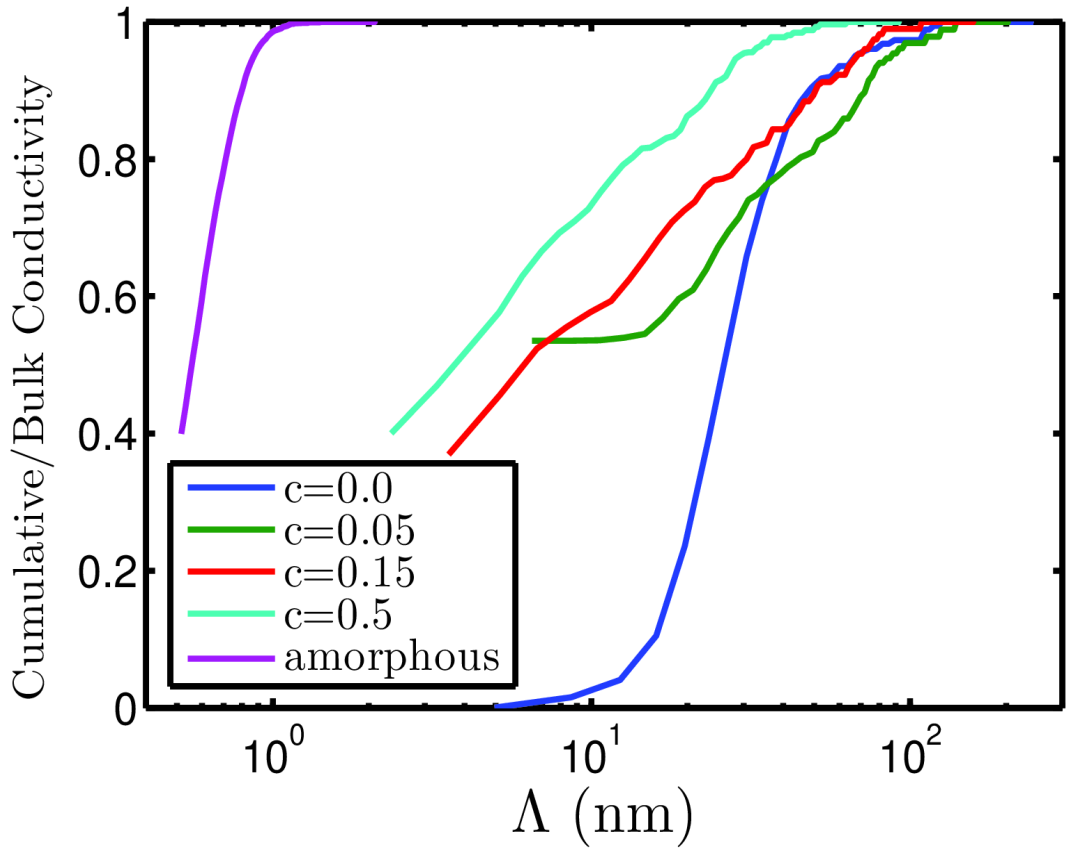


Cumulative Thermal Conductivity

Boundary Scattering: Cumulative Thermal Conductivity

Large c alloys and amorphous vibrations have decreased MFP

Boundary scattering less effective for >10 nm

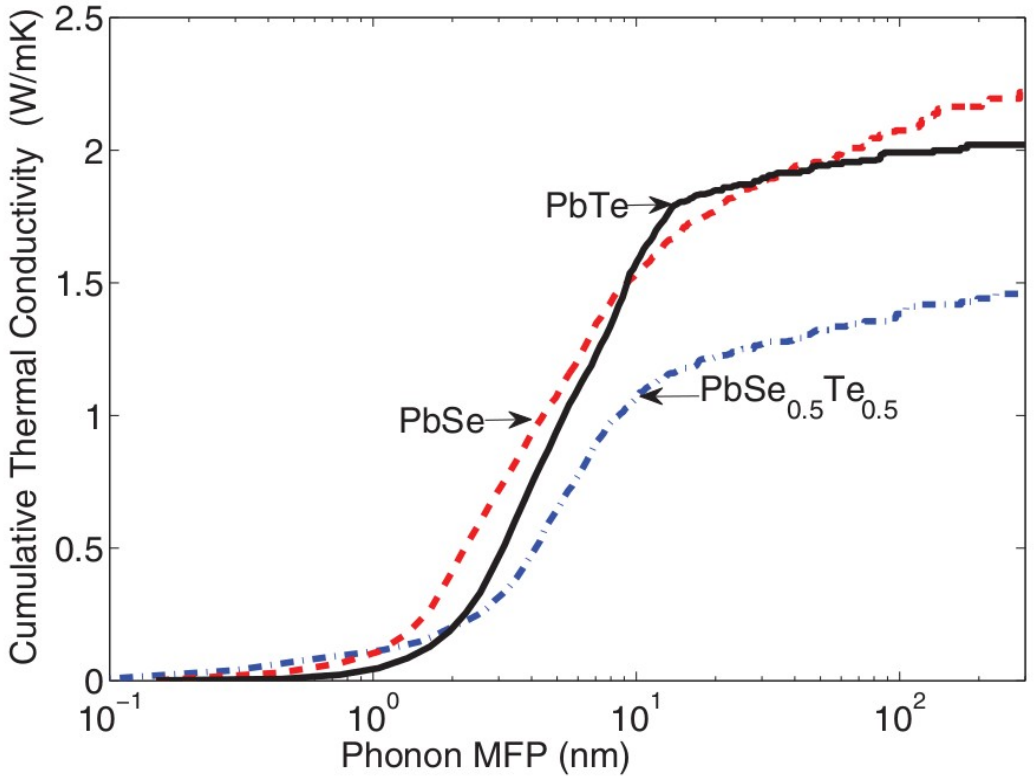


Bulk Thermoelectrics

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



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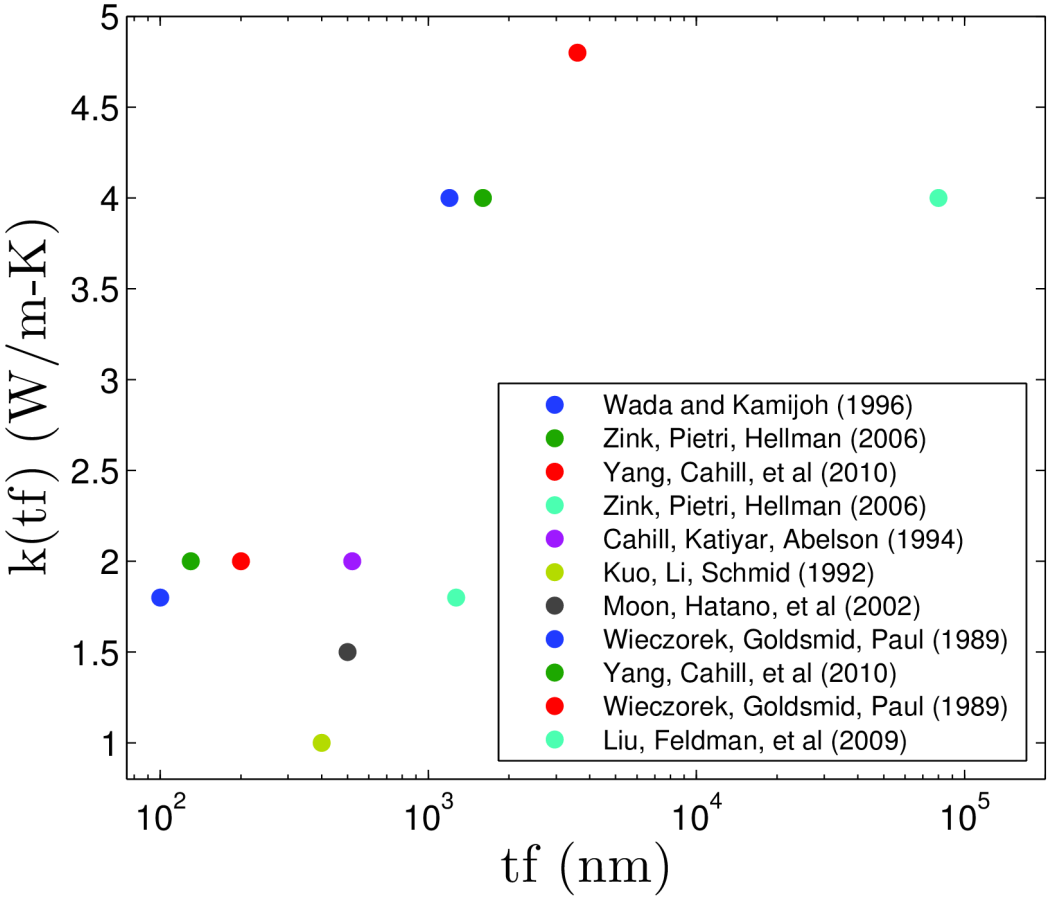
Amorphous Silicon

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

Indicates a phonon-like boundary scattering dependance

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



APPLIED PHYSICS LETTERS 98, 144101 (2011)

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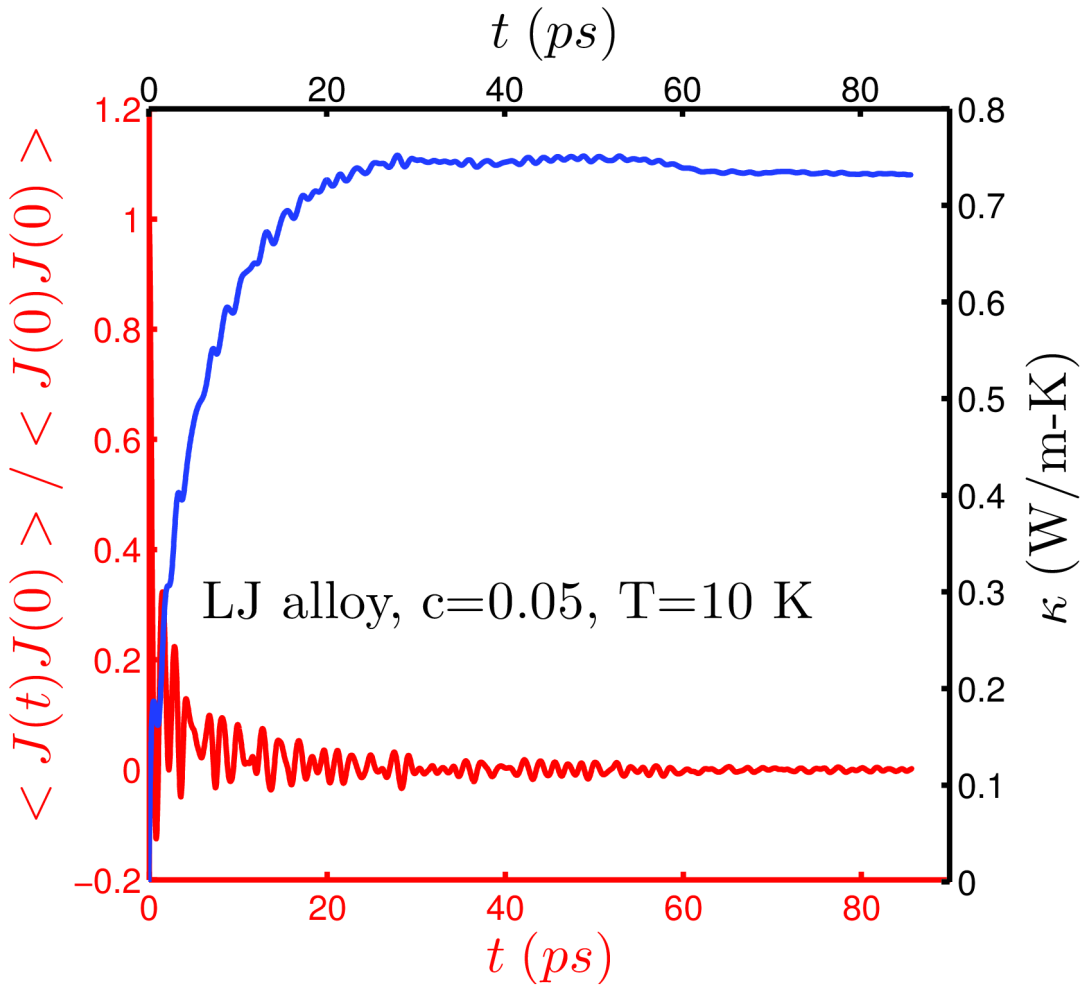
Questions

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.

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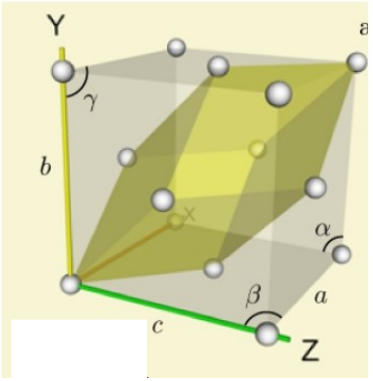
Normal Mode Decomposition (NMD)

Perfect system: vibrations are phonons with an allowed wavevector $\tau\begin{pmatrix}\kappa\\ \nu\end{pmatrix}$



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.

$$\tau\begin{pmatrix}\kappa = 0\\ \omega\end{pmatrix}$$