~~Predicting Phonon Properties of Defected Systems using the Spectral Energy Density~~ Comparison and Evaluation of Spectral Energy Methods for Predicting Phonon Properties

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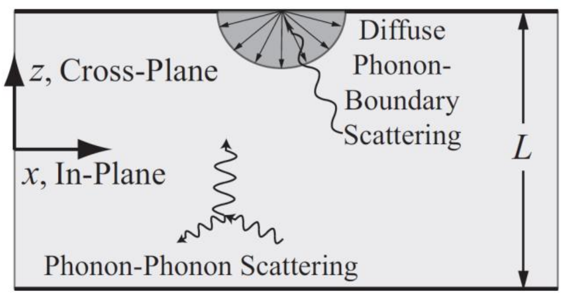
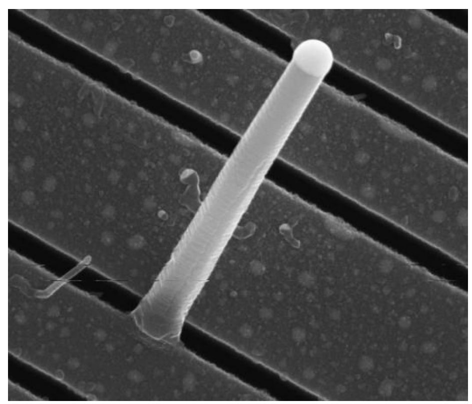
Phonon Properties

-Thermal Conductivity:

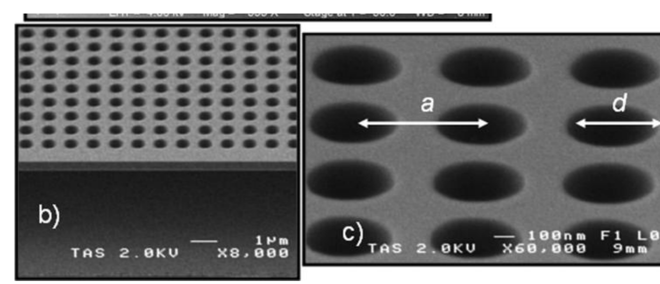
$$k_{\mathbf{n}} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} c\binom{\boldsymbol{\kappa}}{\nu}\, v_{g,\mathbf{n}}^2\binom{\boldsymbol{\kappa}}{\nu} \quad \binom{\boldsymbol{\kappa}}{\nu}$$

-Matthiesen Rule[1,2]:





Nano Letters 9 2009 864-869



Nano Letters **11**, 107-112 (2011).

Accurate phonon lifetimes are important!

[1]A. J. H. McGaughey et al., Applied Physics Letters **99**, 131904 (2011).
[2]D. P. Sellan, et al., Journal of Applied Physics **108**, 113524 (2010).

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Phonon Normal Mode Coordinate[1,2]

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

Need: -**Phonon occupation:** $\begin{pmatrix} \boldsymbol{\kappa} \\ \nu \end{pmatrix}$

1) Allowed wavevectors (from crystal): $\exp[i\boldsymbol{\kappa}\cdot\mathbf{r}_0\left(\begin{smallmatrix} l \\ 0 \end{smallmatrix}\right)]$

2) Atomic velocities (from molecular dynamics): $\dot{u}_\alpha\left(\begin{smallmatrix} l \\ b \end{smallmatrix}; t\right)$

3) Eigenvector (from lattice dynamics): $e^*\left(\begin{smallmatrix} \boldsymbol{\kappa} & b \\ \nu & \alpha \end{smallmatrix}\right)$

- Lattice dynamics requires periodic system.

[1] M. T. Dove, *Introduction to Lattice Dynamics.*
[2] D. C. Wallace, *Thermodynamics of Crystals*.

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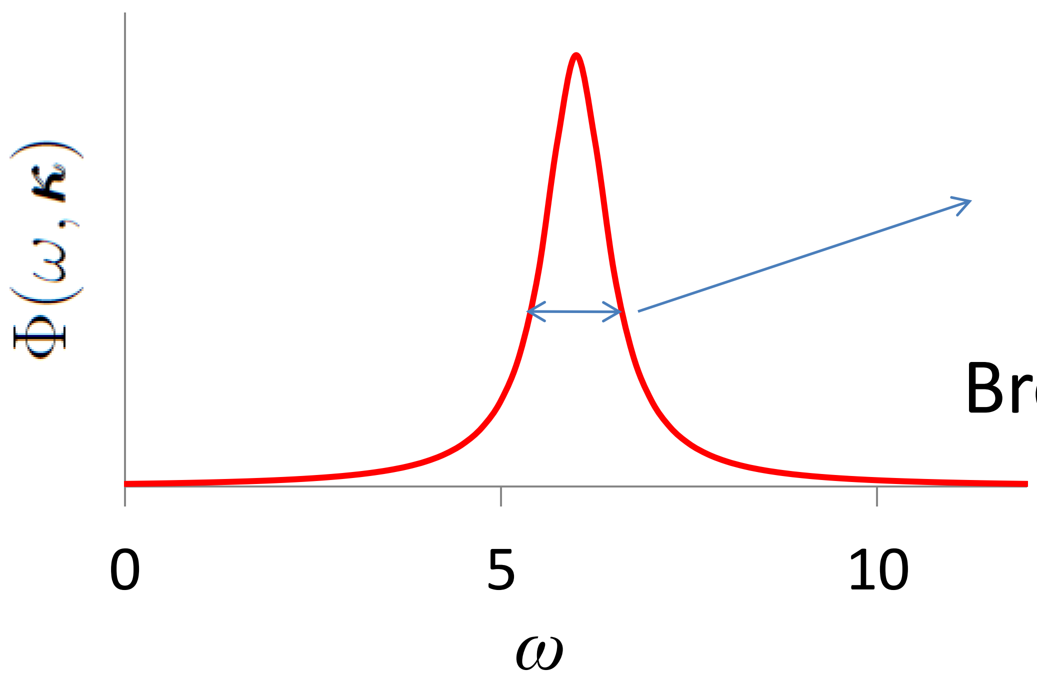
Phonon Lifetime from Spectral Energy

$$\Phi(\omega, \boldsymbol{\kappa}) = \lim_{\tau_0 \to \infty} \frac{1}{2\tau_0} \left| \frac{1}{\sqrt{2\pi}} \int_0^{\tau_0} \dot{q}\binom{\boldsymbol{\kappa}}{\nu}; t) \exp(-i\omega t)\, dt \right|^2$$

$$= \sum_{\nu}^{3n} C_0\binom{\boldsymbol{\kappa}}{\nu} \frac{\Gamma\binom{\boldsymbol{\kappa}}{\nu} / \pi}{\left[\omega_0\binom{\boldsymbol{\kappa}}{\nu} - \omega\right]^2 + \Gamma^2\binom{\boldsymbol{\kappa}}{\nu}}$$



$$\binom{\boldsymbol{\kappa}}{\nu} = 1/2\Gamma\binom{\boldsymbol{\kappa}}{\nu}$$

Broad peak = short **lifetime**

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Proposed Phonon Spectral Energy[1,2]

$$\Phi'(\omega,\boldsymbol{\kappa}) = \frac{1}{4\pi\tau_0}\sum_{\alpha}^{3}\sum_{b}^{n}\frac{m_b}{N}\left|\sum_{l}^{N}\int_0^{\tau_0}\dot{u}_\alpha\left(\begin{smallmatrix}l\\b\end{smallmatrix};t\right)\exp[i\boldsymbol{\kappa}\cdot\mathbf{r}_0\left(\begin{smallmatrix}l\\0\end{smallmatrix}\right)-i\omega t]dt\right|^2$$

Need:

1) Allowed wavevectors: $\exp[i\boldsymbol{\kappa}\cdot\mathbf{r}_0\left(\begin{smallmatrix}l\\0\end{smallmatrix}\right)]$

2) Atomic velocities: $\dot{u}_\alpha\left(\begin{smallmatrix}l\\b\end{smallmatrix};t\right)$

3) ~~Eigenvector from lattice dynamics:~~ $\cancel{e^*\left(\begin{smallmatrix}\boldsymbol{\kappa} & b\\ \nu & \alpha\end{smallmatrix}\right)}$

- Measure lifetimes in **defected/disordered** systems!
- Does NOT represent phonon spectral energy!

[1]S. Maruyama, Microscale Thermophysical Engineering **7**, 41 (2003).
[2]J. A. Thomas, et al., Physical Review B **81**, 081411(R) (2010).

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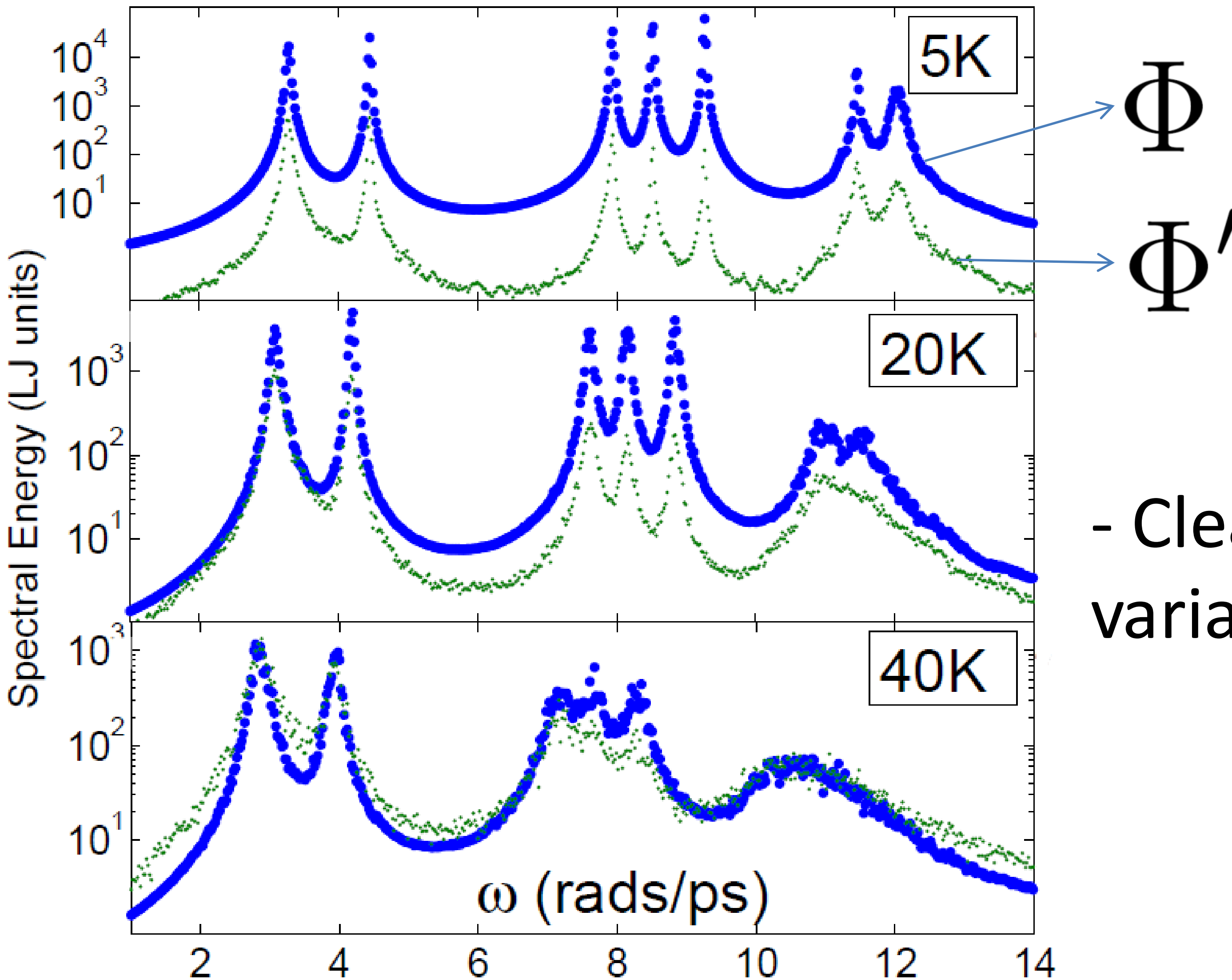
Phonon Spectral Energy Comparison

K = [π/2a 00]



- Clear linewidth variations:

$$\binom{\boldsymbol{\kappa}}{\nu} = 1/2\Gamma\binom{\boldsymbol{\kappa}}{\nu}$$

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3 Case Studies

- **Lennard-Jones Ar**: well-studied, computationally cheap

- **Stillinger-Weber Silicon**[1]: "stiff" system (high frequencies, large group velocities, long lifetimes).

- **Carbon nanotube using REBO**[2,3]: test/validate previous work.

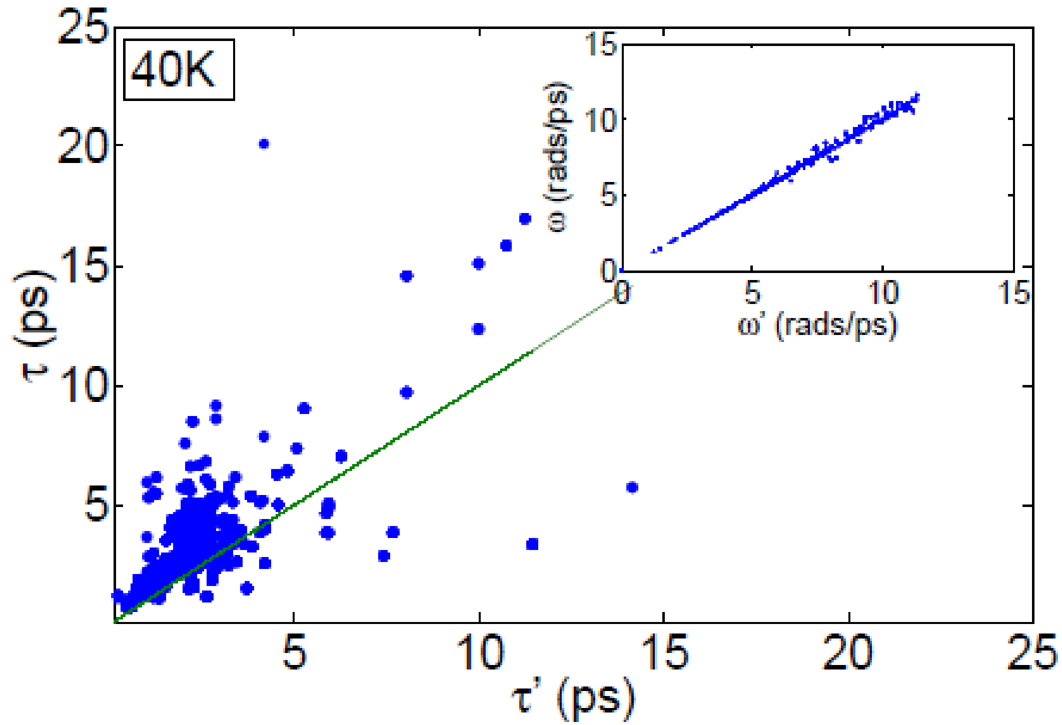
[1]F. H. Stillinger and T. A. Weber, Physical Review B **31**, 5262 (1985).

[2]D. W. Brenner, et al., Journal of Physics: Condensed Matter **14**, 783 (2002).

[3]J. A. Thomas, et al., Physical Review B **81**, 081411(R) (2010).

Case Study: LJ Lifetimes

$\Phi \; \tau \; \omega$



$\Phi' \; \tau' \; \omega'$

-Frequencies agree well, increasing scatter with T(K)

-Lifetimes large scatter independent of T(K)

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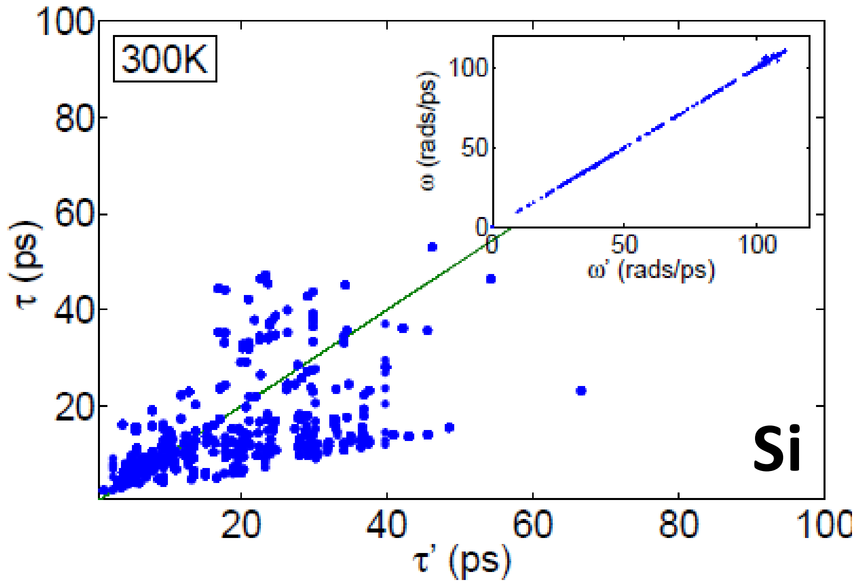
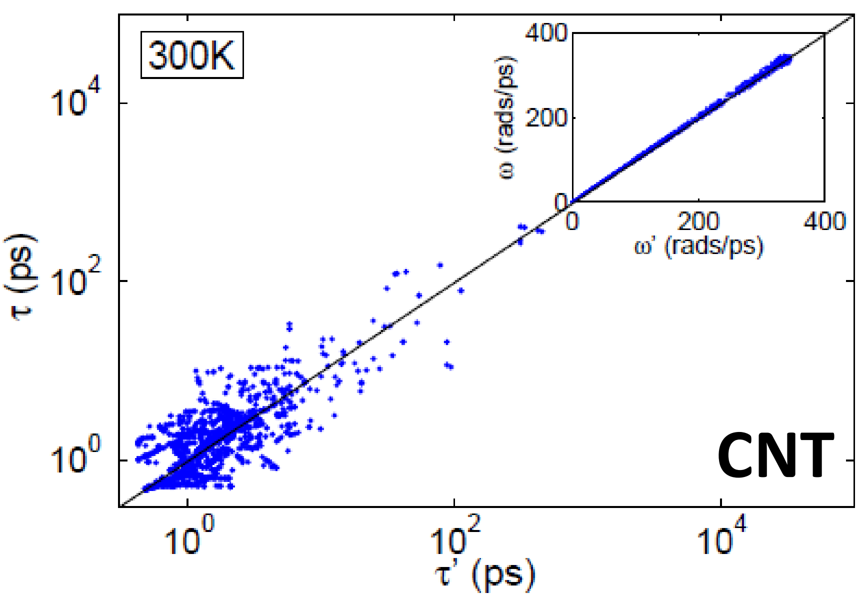
Case Study: SW Si and CNT Lifetimes



-Frequencies agree well
-Lifetimes large scatter

$\Phi' \tau' \omega' \quad \Phi \, \tau \, \omega$



-Frequencies agree well
-Lifetimes large scatter

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Thermal Conductivity Predictions: LJ

TABLE I: Thermal conductivity values in W/m-K predicted using Φ, Φ', and the Green-Kubo

T (K)	GK-MD	Φ	Φ'
LJ			
5	8.0	7.9	5.8
20	1.3	1.2	1.0
40	0.45	0.47	0.49

- LJ results for Φ and other methods agree[1], Φ' does not.
- Results from exact same molecular dynamics data.

[1] J. E. Turney, et al., Physical Review B **79**, 064301 (2009).

[2] J. A. Thomas, et al., Physical Review B **81**, 081411(R) (2010).

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Thermal Conductivity Predictions: SW Si and CNT

TABLE I: Thermal conductivity values in W/m-K predicted using Φ, Φ', and the Green-Kubo

T (K)	GK-MD	Φ	Φ'
SW (N_0=6)			
300		530	651
CNT (N_0=50)			
300		428	398

- SW Si and CNT[2] results indicate no systematic behavior for Φ'.

[1]J. E. Turney, et al., Physical Review B **79**, 064301 (2009).

[2]J. A. Thomas, et al., Physical Review B **81**, 081411(R) (2010).

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Proposed Phonon Spectral Energy Density

$$\Phi'(\omega,\boldsymbol{\kappa}) = \frac{1}{4\pi\tau_0}\sum_{\alpha}^{3}\sum_{b}^{n}\frac{m_b}{N}\left|\sum_{l}^{N}\int_0^{\tau_0}\dot{u}_\alpha\left({l \atop b};t\right)\exp[i\boldsymbol{\kappa}\cdot\mathbf{r}_0\left({l \atop 0}\right)-i\omega t]dt\right|^2$$

- Should **NOT** be used to predict lifetimes or thermal conductivity!

- Can accurately measure frequencies, even in **disordered** systems.

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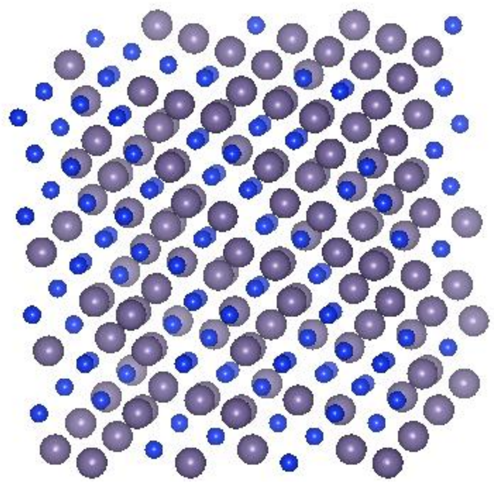
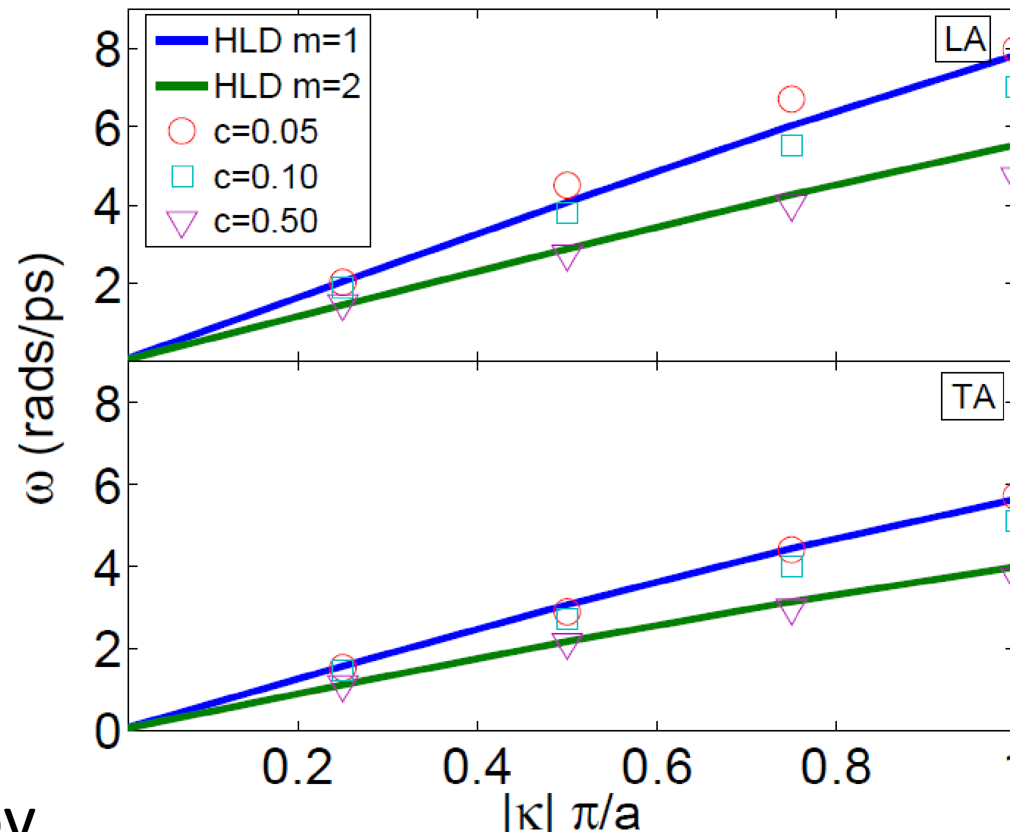
Dispersion in Disordered Systems



$c=0.5$

$m_e=2$



- LJ alloys with:

$m_a = 1$

$m_b = 3$

- Dispersion well approx. by lattice dynamics w/m_e

- Original purpose for Φ' [1]!

[1] S. Maruyama, Microscale Thermophysical Engineering **7**, 41 (2003).

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Questions

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This work is supported by AFOSR award FA95501010098.

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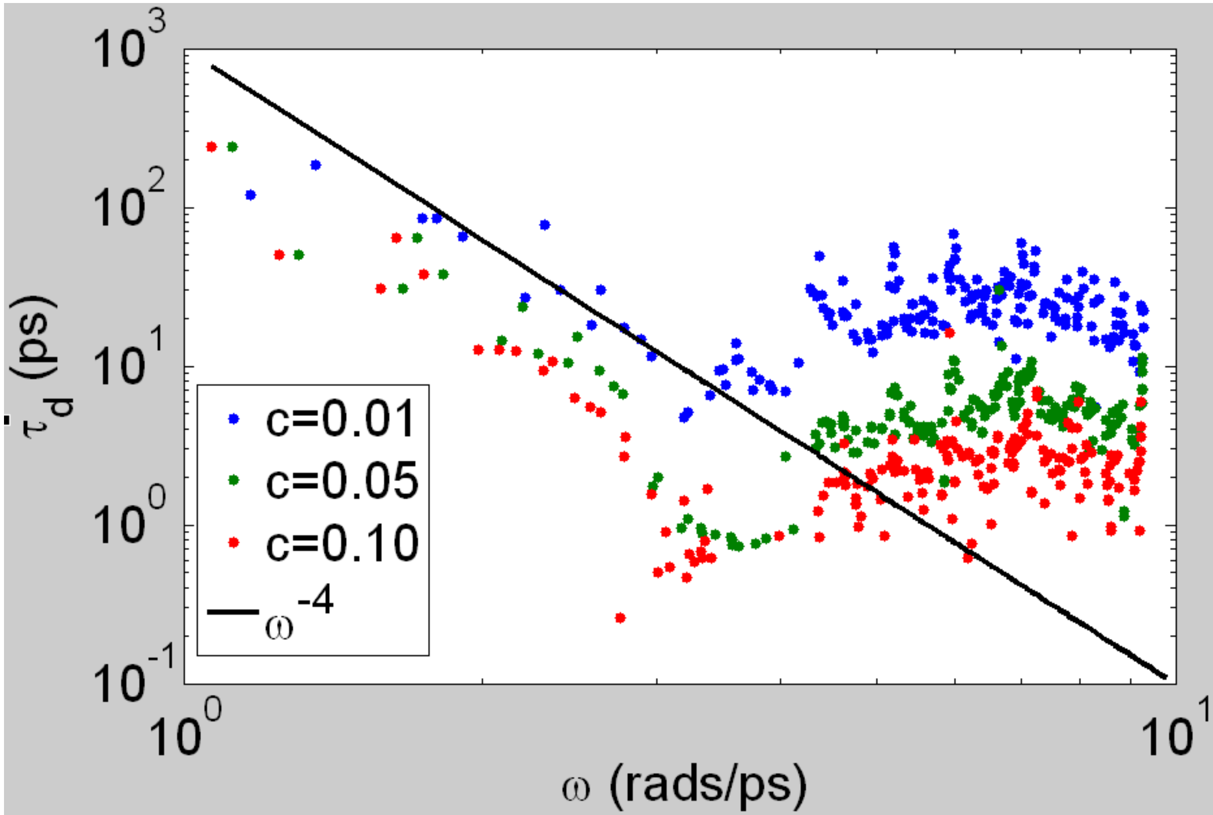
Defect Scattering



-Matthiesen Rule:

-Rayleigh scattering:

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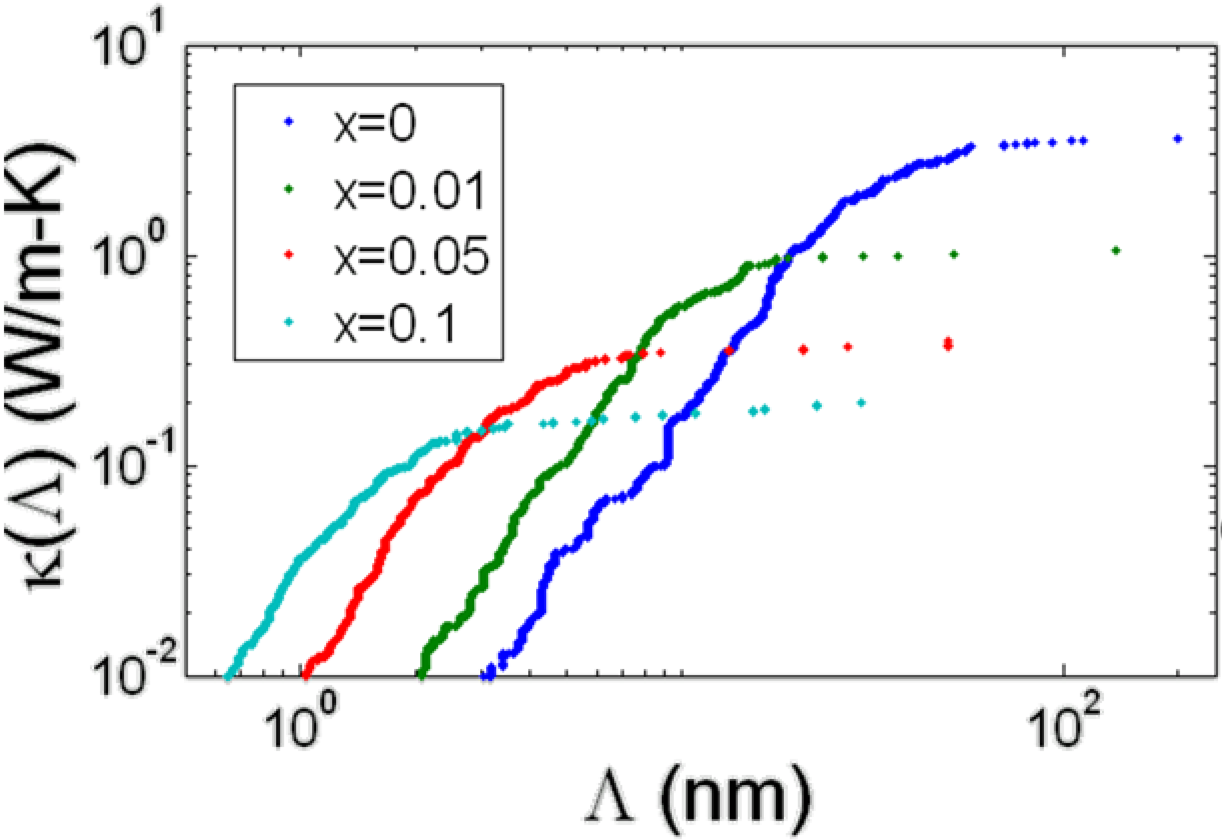
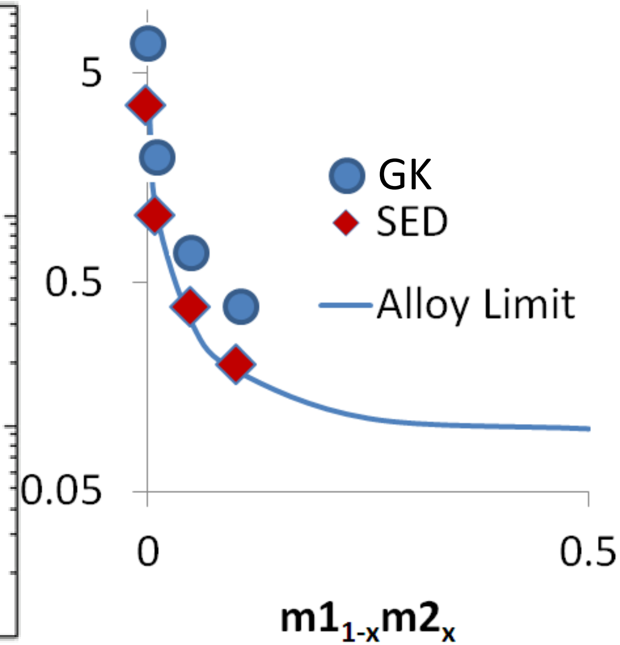
Thermal Conductivity of Thin Films





$$L \approx \Lambda$$

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