Evaluation of the Virtual Crystal Approximation for Predicting Alloy Vibrational Mode Properties and Thermal Conductivity

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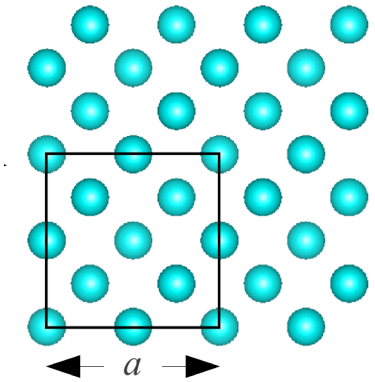
http://ntpl.me.cmu.edu/

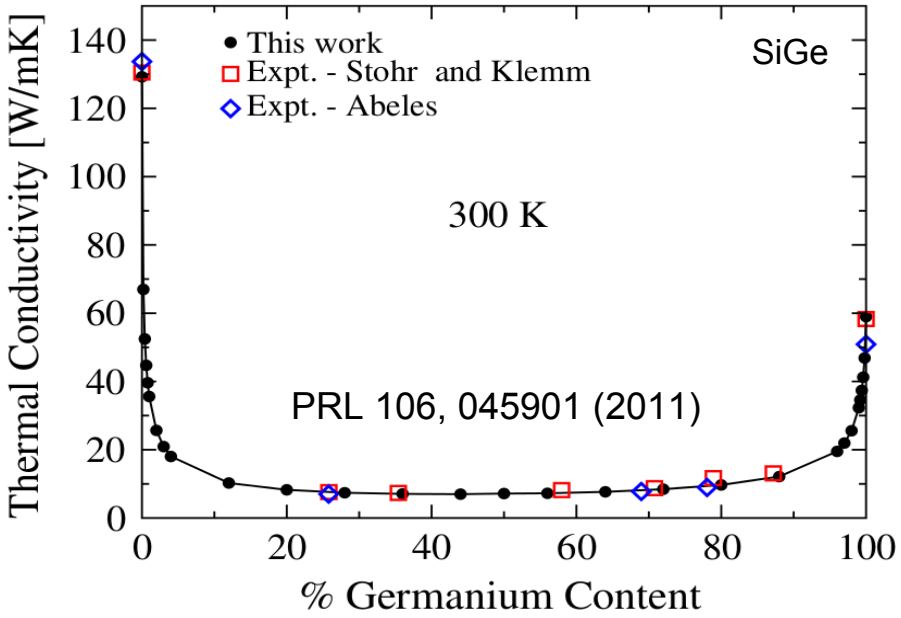
04/04/13

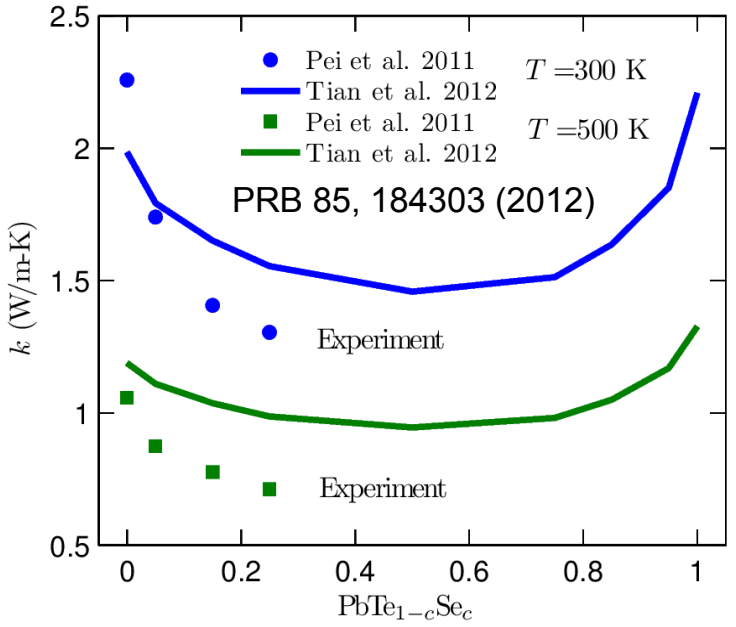
Motivation: experimental accuracy

Density Functional Theory (DFT) + **(VC-ALD)**

(VC-ALD) = **(VC)** Virtual Crystal approximation
+
(ALD) Anharmonic Lattice Dynamics



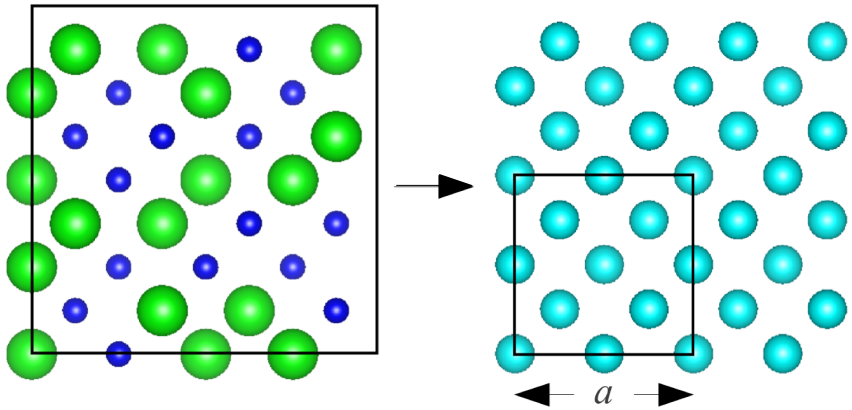




Virtual Crystal Approximation

Gamma

VC



$$\bar{m}^{\mu} = (1-c)m^{i} + cm^{j}$$

$$k_{ph,\mathbf{n}} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} \frac{k_B}{V} D_{ph,\mathbf{n}}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right)$$

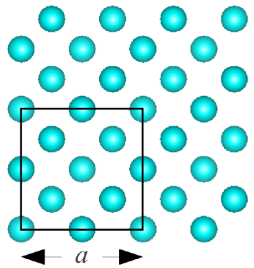
$$D_{ph,\mathbf{n}}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right) = v^2_{g,\mathbf{n}}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right) \tau\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix}\right)$$

B. Abeles, Phys. Rev. 131, 19061911 (1963)

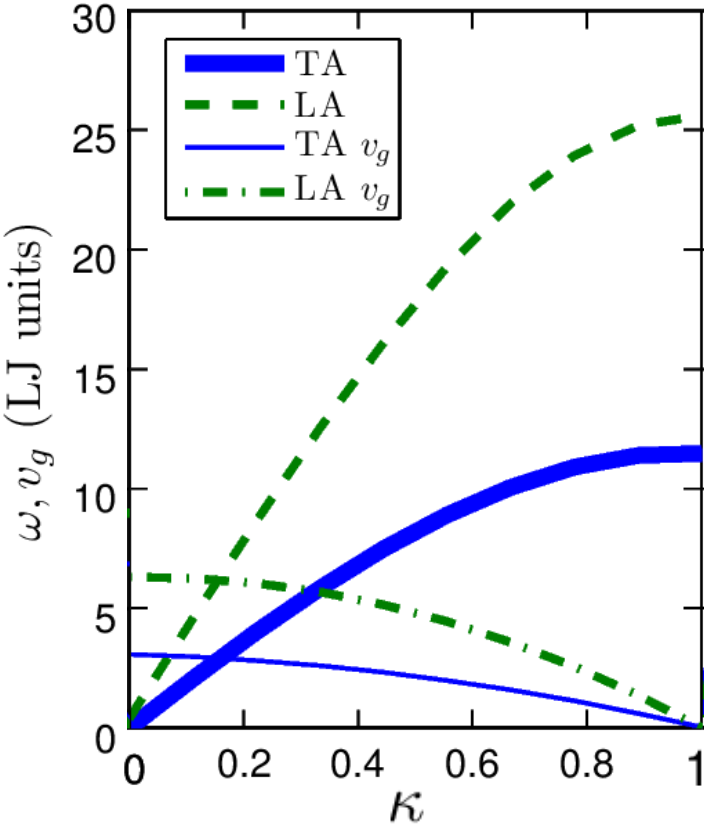
VC-ALD Diffusivities

$$D_{ph,\mathbf{n}}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right) = v^2_{g,\mathbf{n}}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)\tau\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)$$

VC Unit Cell



$$v_{g,\mathbf{n}}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right) = \frac{\partial\omega\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)}{\partial\boldsymbol{\kappa}}$$

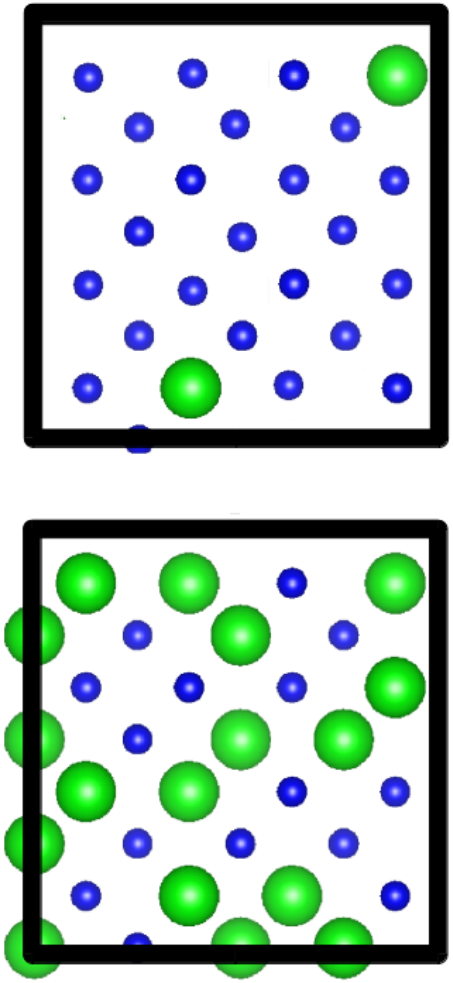


Matthiessen's Rule

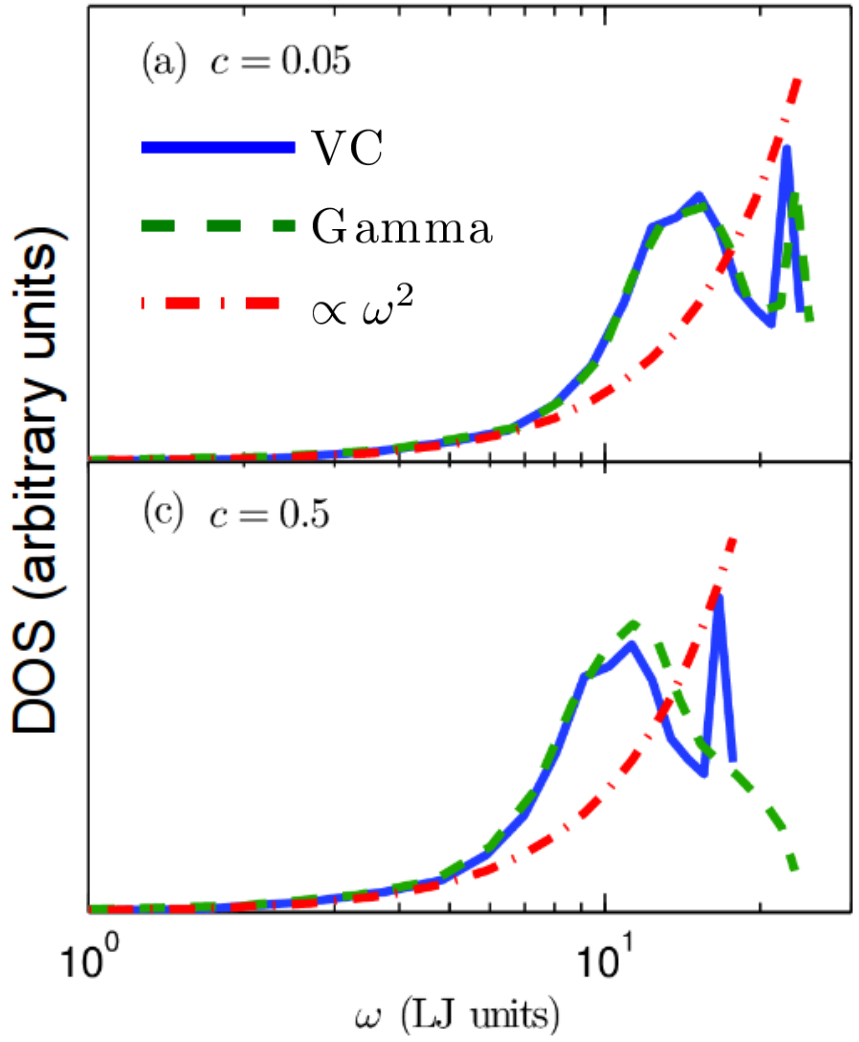
$$\frac{1}{\tau\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)} = \frac{1}{\tau_{p-p}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)} + \frac{1}{\tau_{p-d}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)}$$

Explicit disorder: VC vs Gamma

Gamma



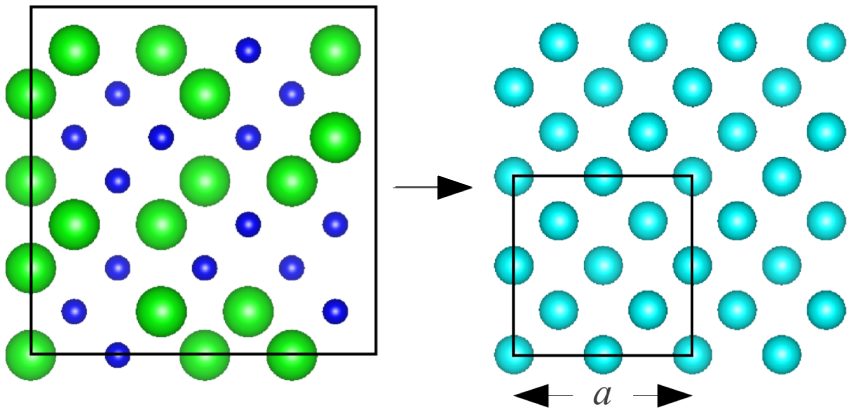
Lennard-Jones Argon Alloys



Explicit disorder: Structure Factor

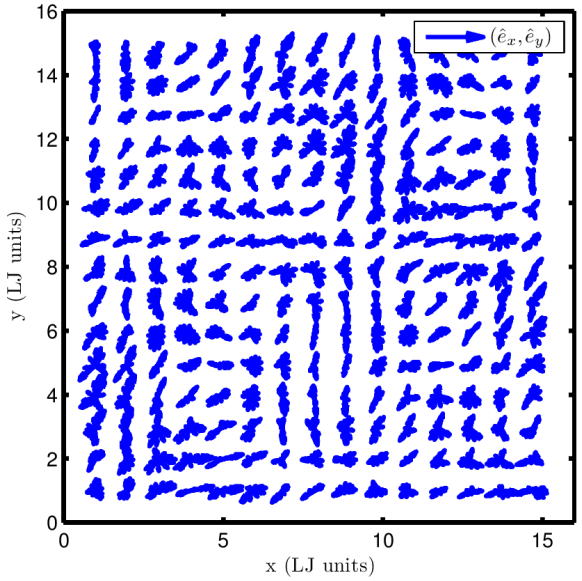
Gamma

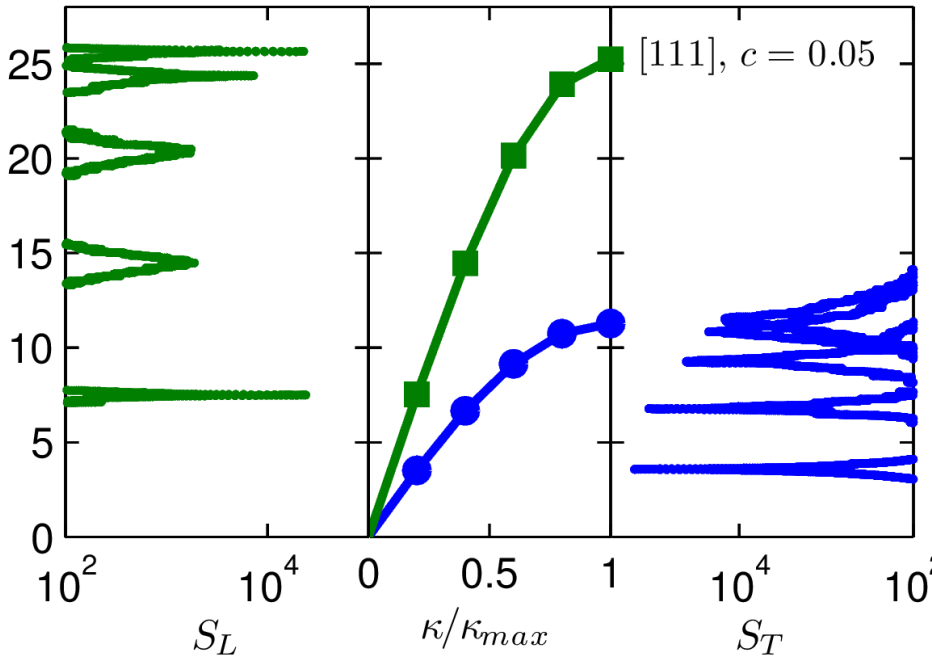
VC



$$D_{ph,\mathbf{n}}(\kappa_\nu) = v^2_{g,\mathbf{n}}(\kappa_\nu)\,\tau(\kappa_\nu)$$

Gamma VC Gamma

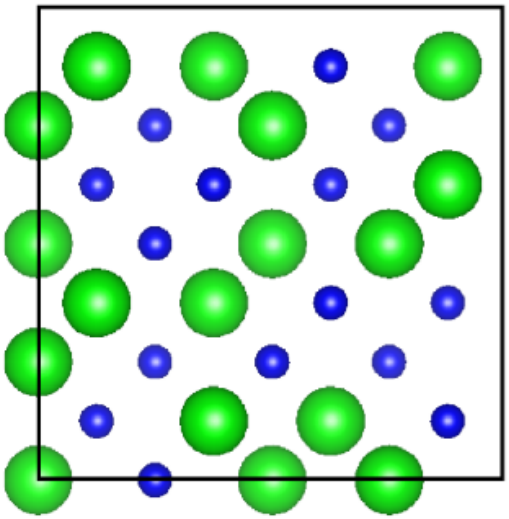




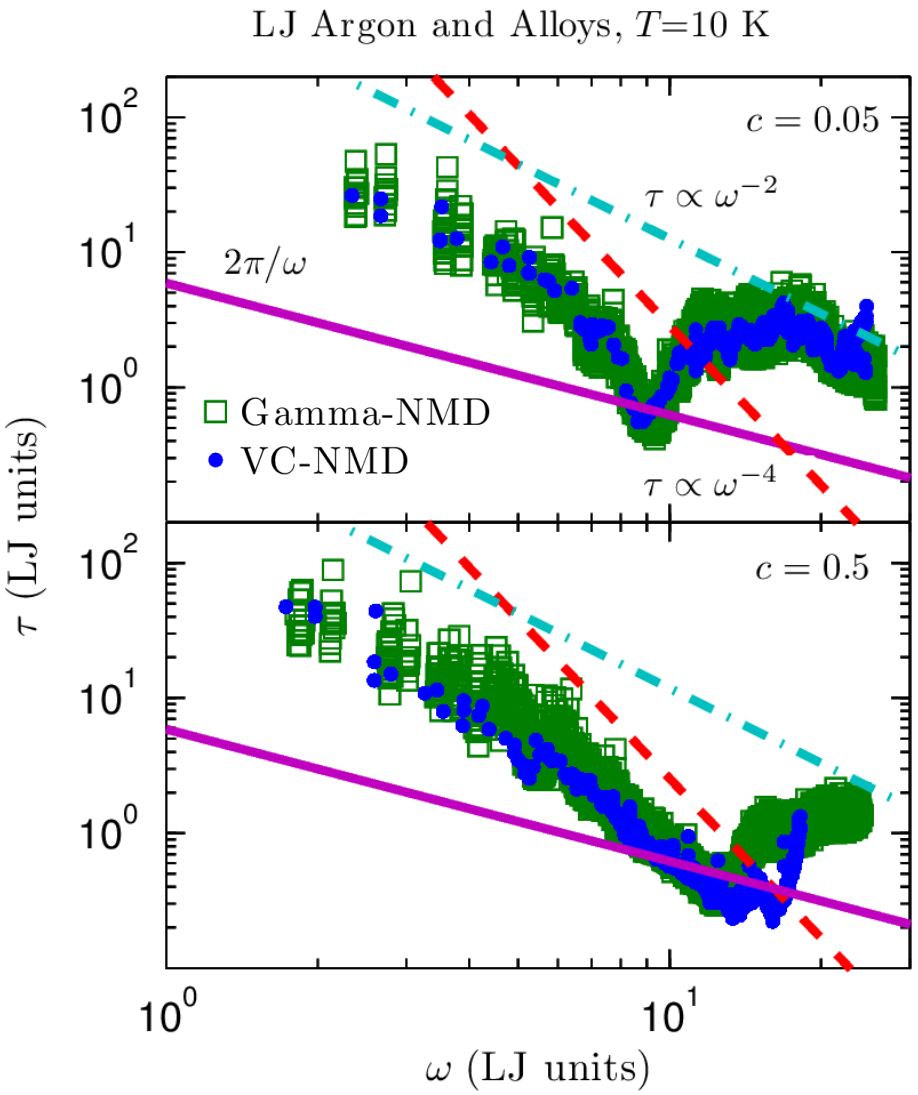
Phys. Rev. B 59, 35513559 (1999)

Normal Mode Decomposition (NMD)

Molecular Dynamics Gamma



$$\tau(\kappa_\nu) = \int_0^{t^*} \frac{< E(\kappa_\nu; t)\, E(\kappa_\nu; 0) >}{< E(\kappa_\nu; 0)\, E(\kappa_\nu; 0) >} dt$$

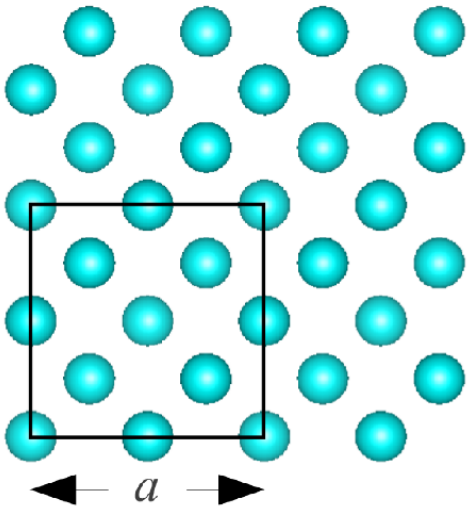


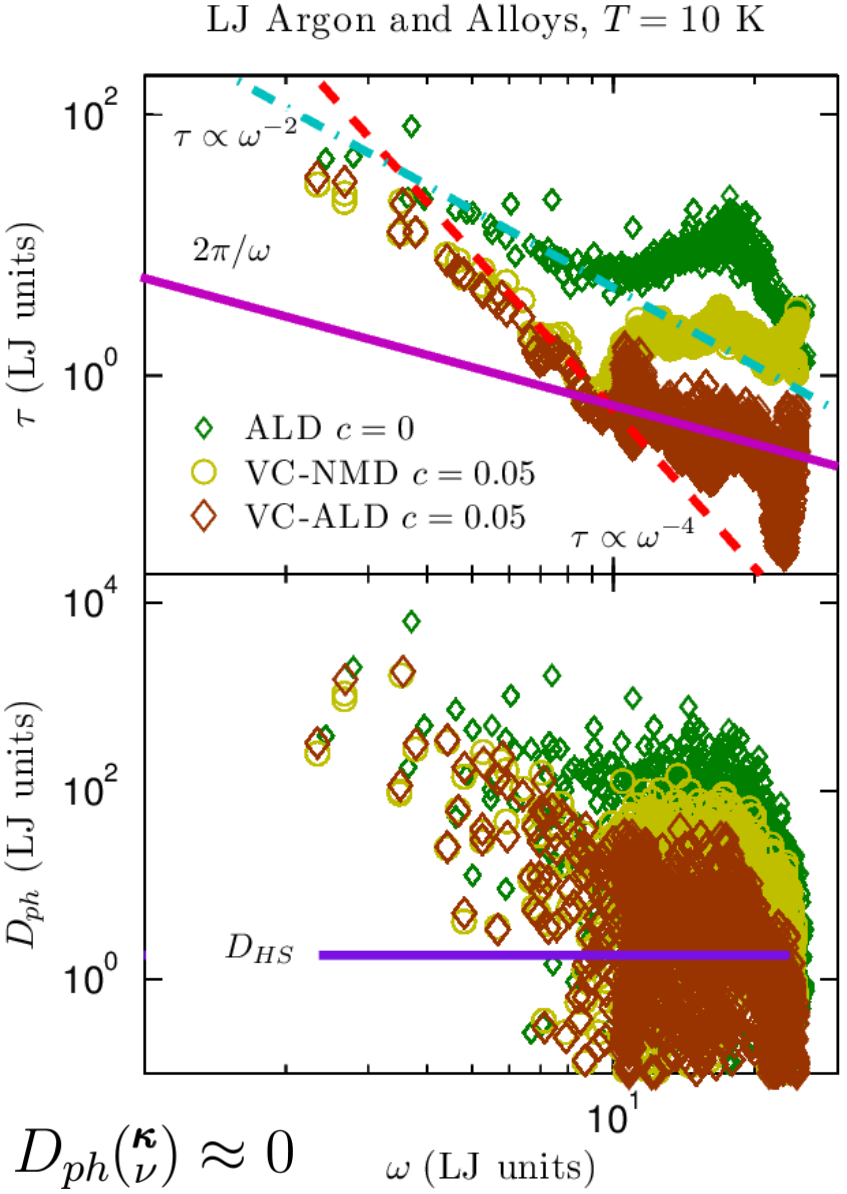
VC Diffusivities

$$D_{ph,\mathbf{n}}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right) = v^2_{g,\mathbf{n}}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)\tau\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)$$

$$D_{HS} = \frac{1}{3} v_s a$$

VC Unit Cell

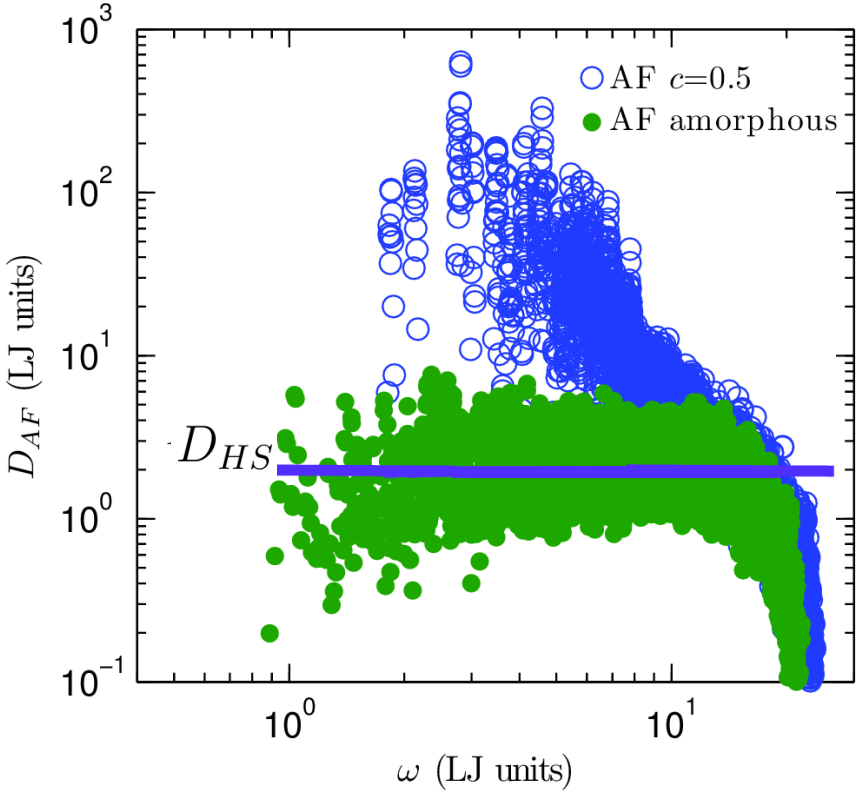


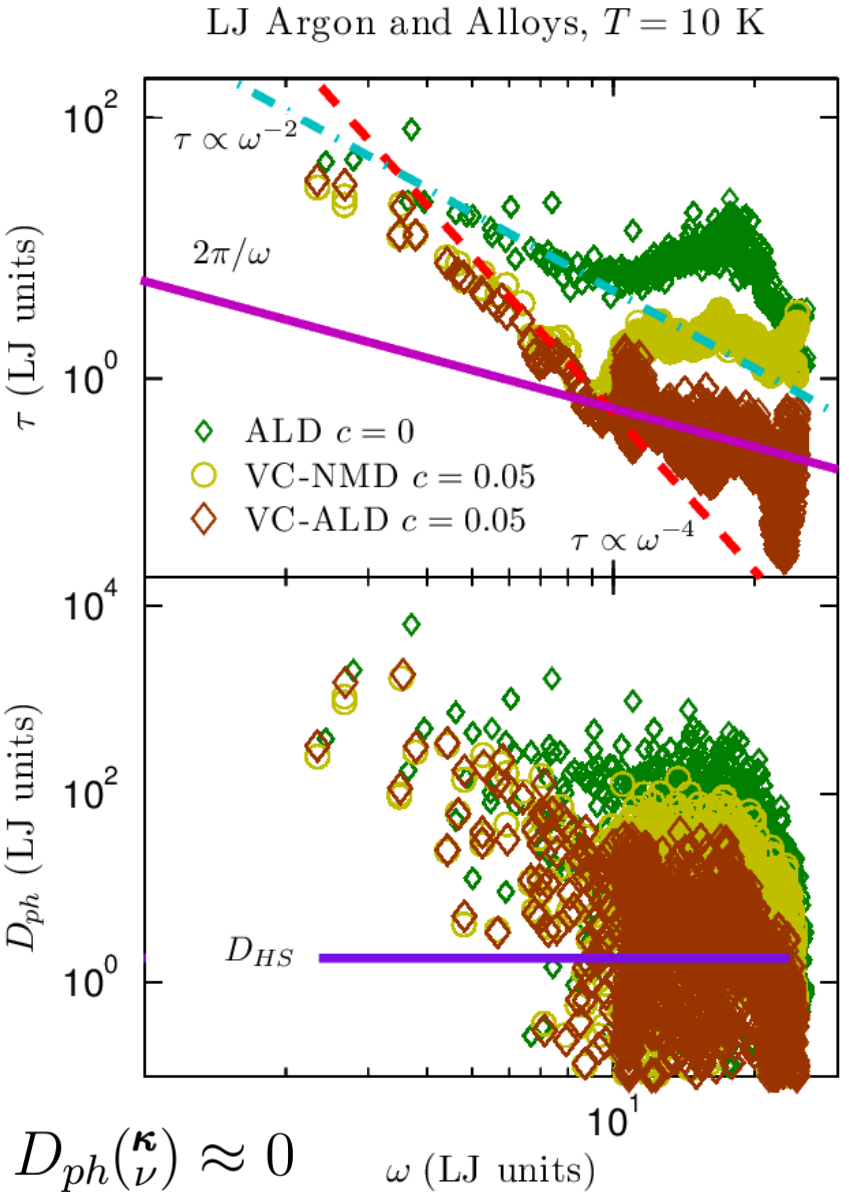


$$D_{ph}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right) \approx 0$$

AF Diffusivities

$$k_{AF} = \sum_{diffusons} \frac{k_B}{V} D_{AF,i}(\omega_i)$$





$D_{ph}(\kappa_\nu) \approx 0$

Physical Review B 48, 1258112588 (1993)

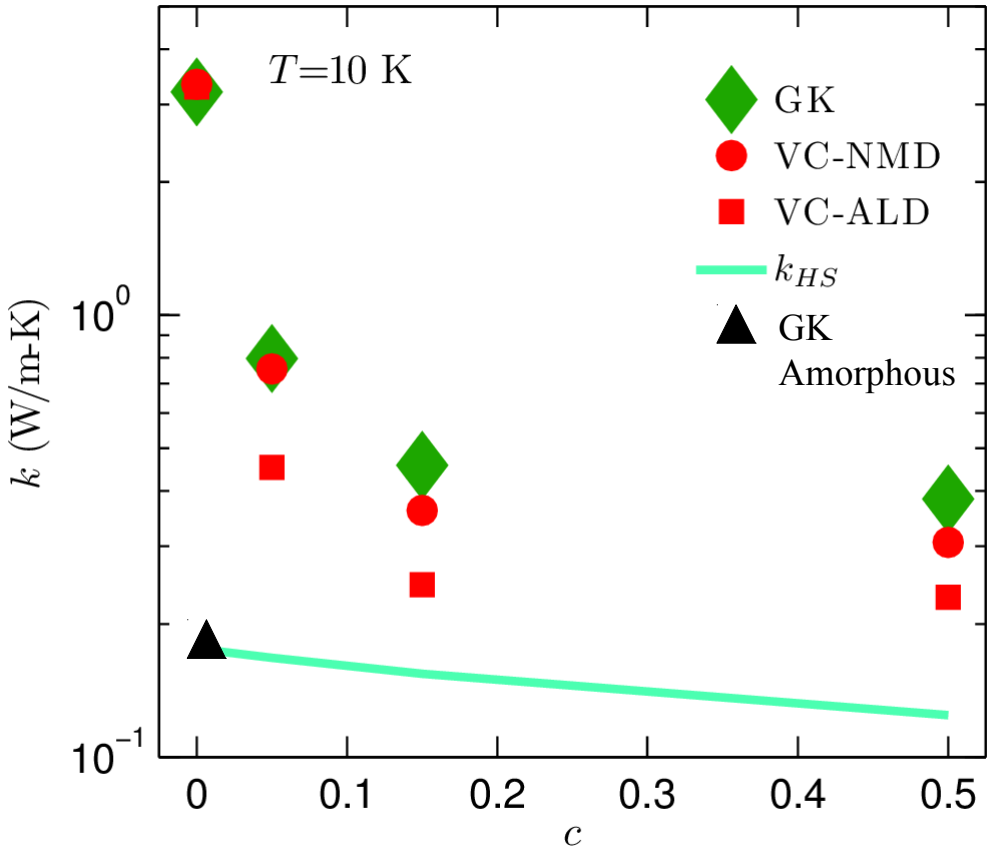
Thermal conductivity

Green-Kubo (GK): MD-based, no assumptions

High-scatter adjustment*:

$$D_{ph}\left(\begin{matrix}\kappa\\ \nu\end{matrix}\right) < D_{HS}$$

$$D_{ph}\left(\begin{matrix}\kappa\\ \nu\end{matrix}\right) = D_{HS}$$



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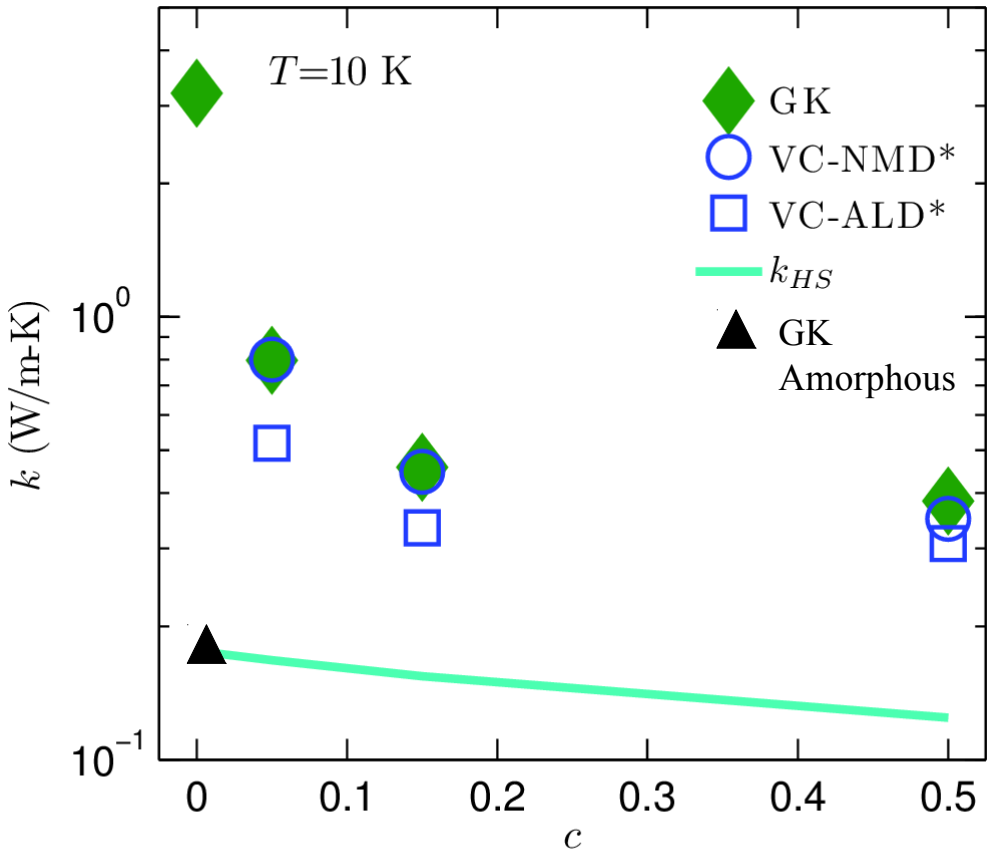
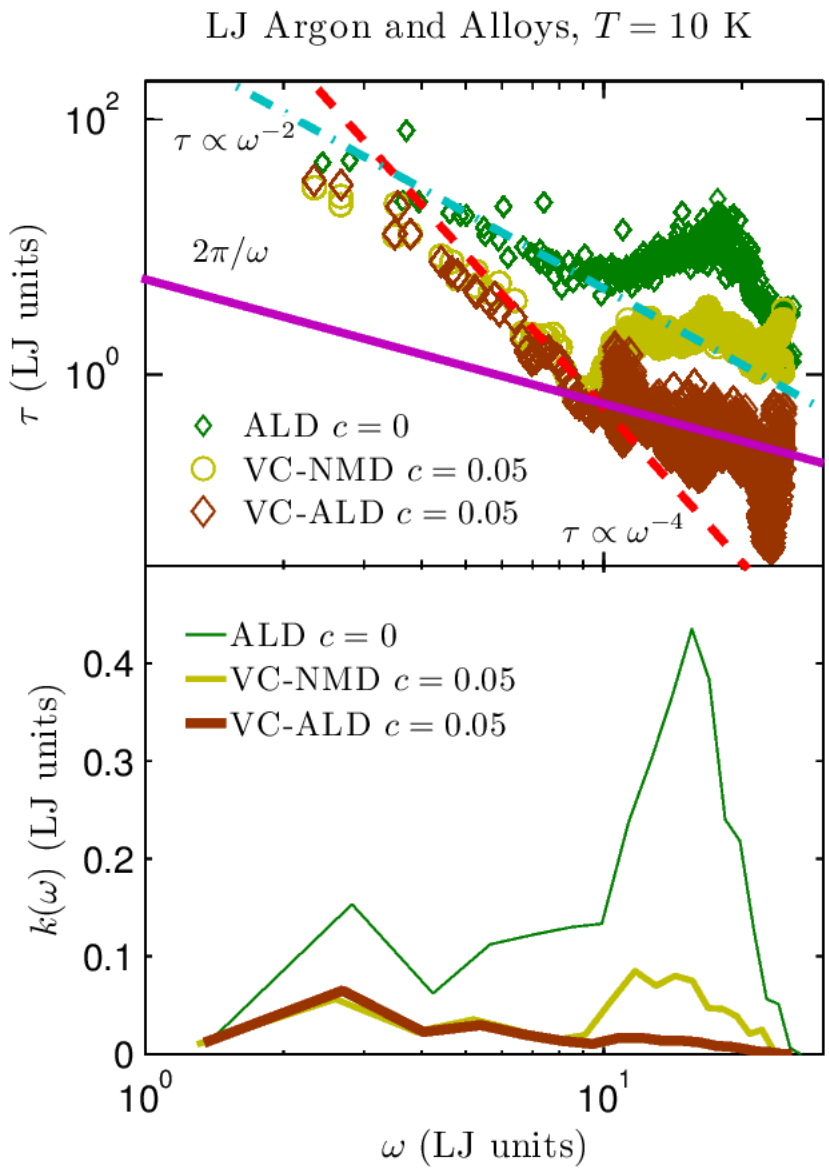
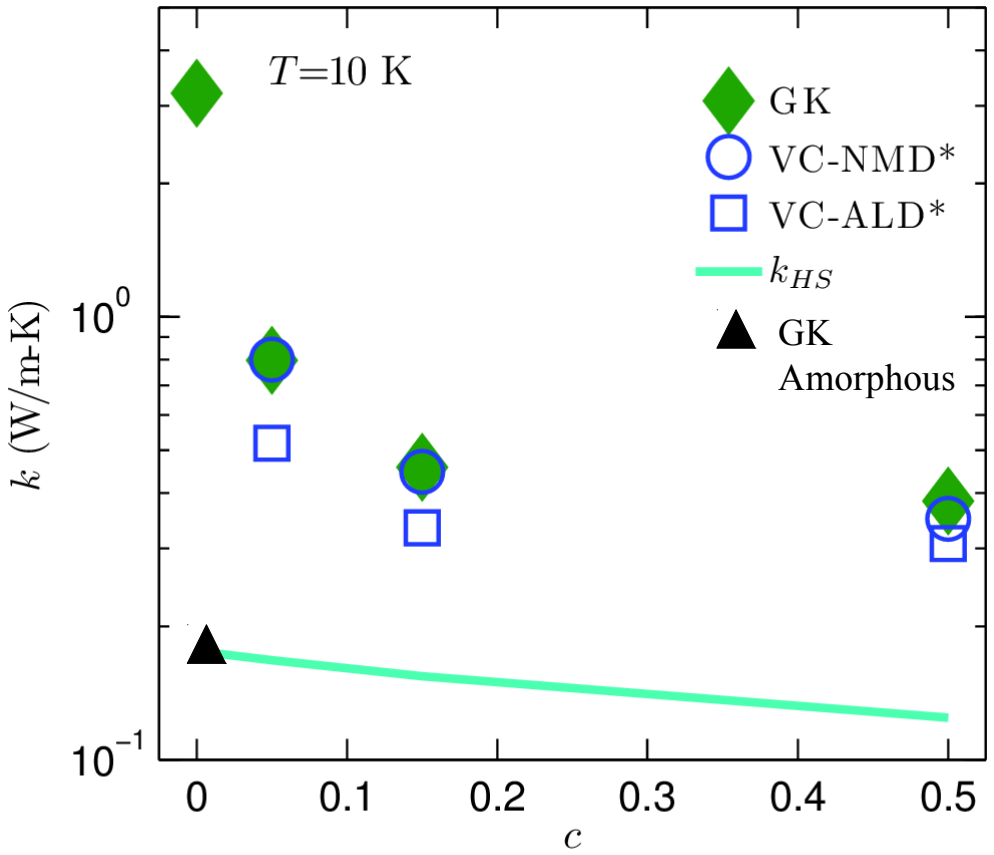


Thermal conductivity spectrum





Summary

VC approximation underpredicts mode group velocities at high frequency, which lead to underprediction of mode diffusivity.

VC-ALD underpredicts lifetimes at high-frequency.

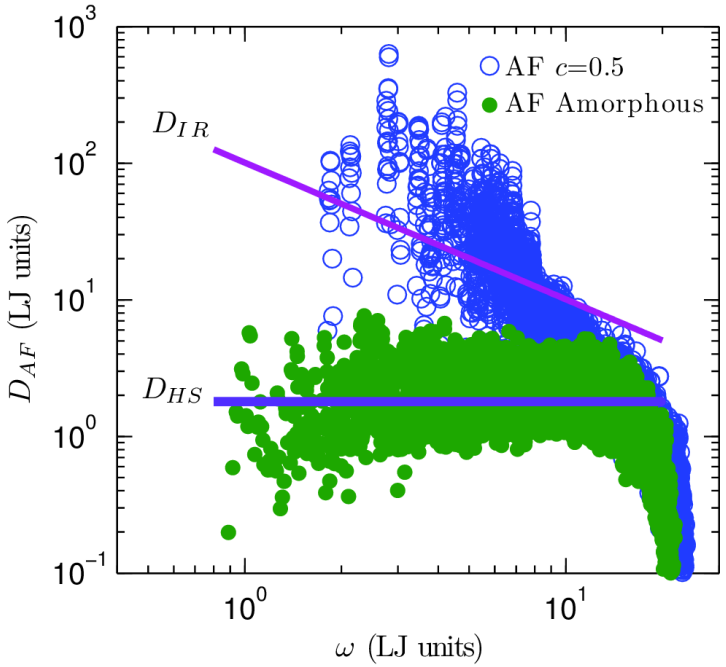
LJ Argon has important contribution from high-frequency modes to thermal conductivity.

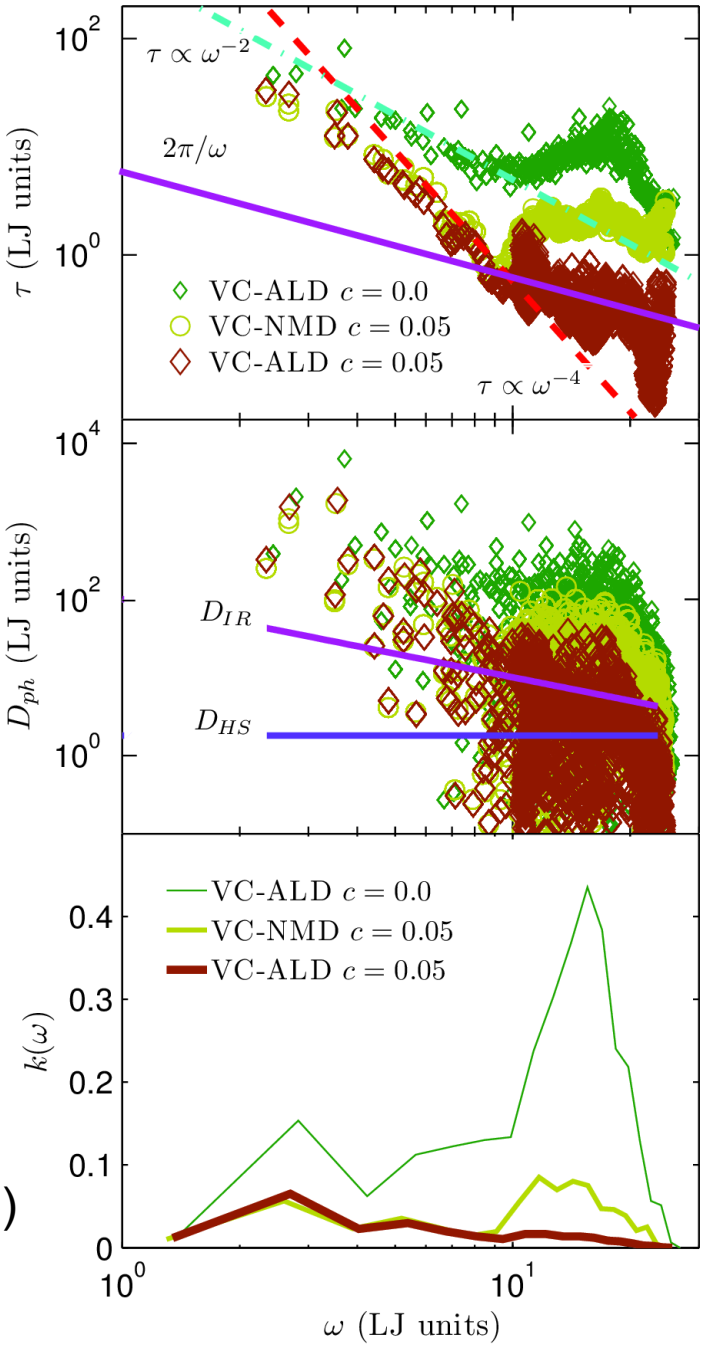
Breakdown of VC-ALD method is likely for materials with thermal conductivity near the high-scatter limit, or for modes below the high-scatter limit.

This work was supported by AFOSR award FA95501010098 and by a grant of computer time from the DOD High Performance Computing Modernization Program at the US Army Engineer Research and Development Center. We thank Davide Donadio, Jivtesh Garg, Asad Hasan, Craig Maloney, and Zhiting Tian for helpful discussions.

HS/IR Limit

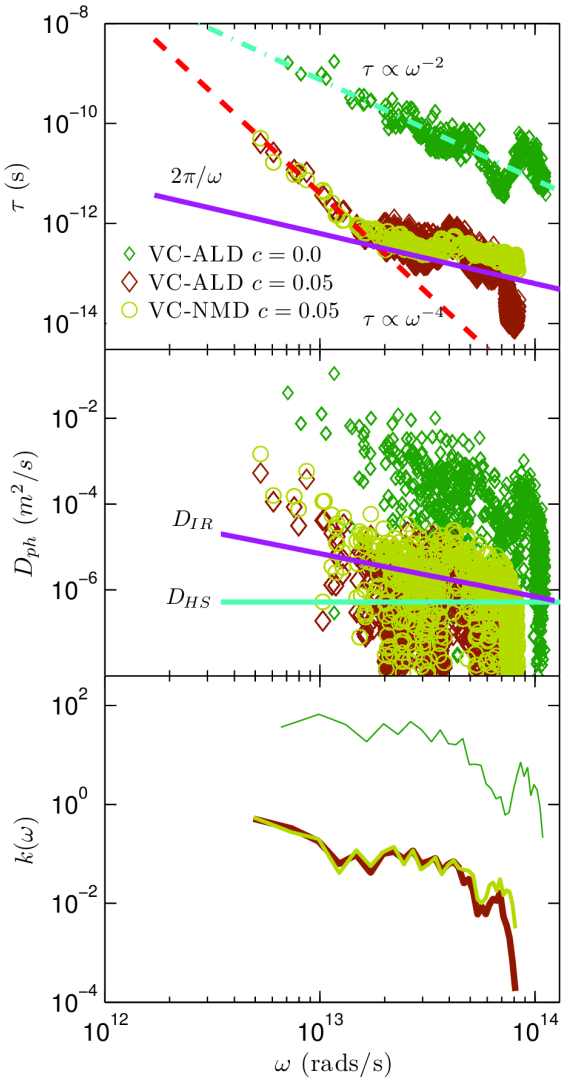
$$D_{IR} = \frac{2\pi}{3}\frac{v_s^2}{\omega}$$

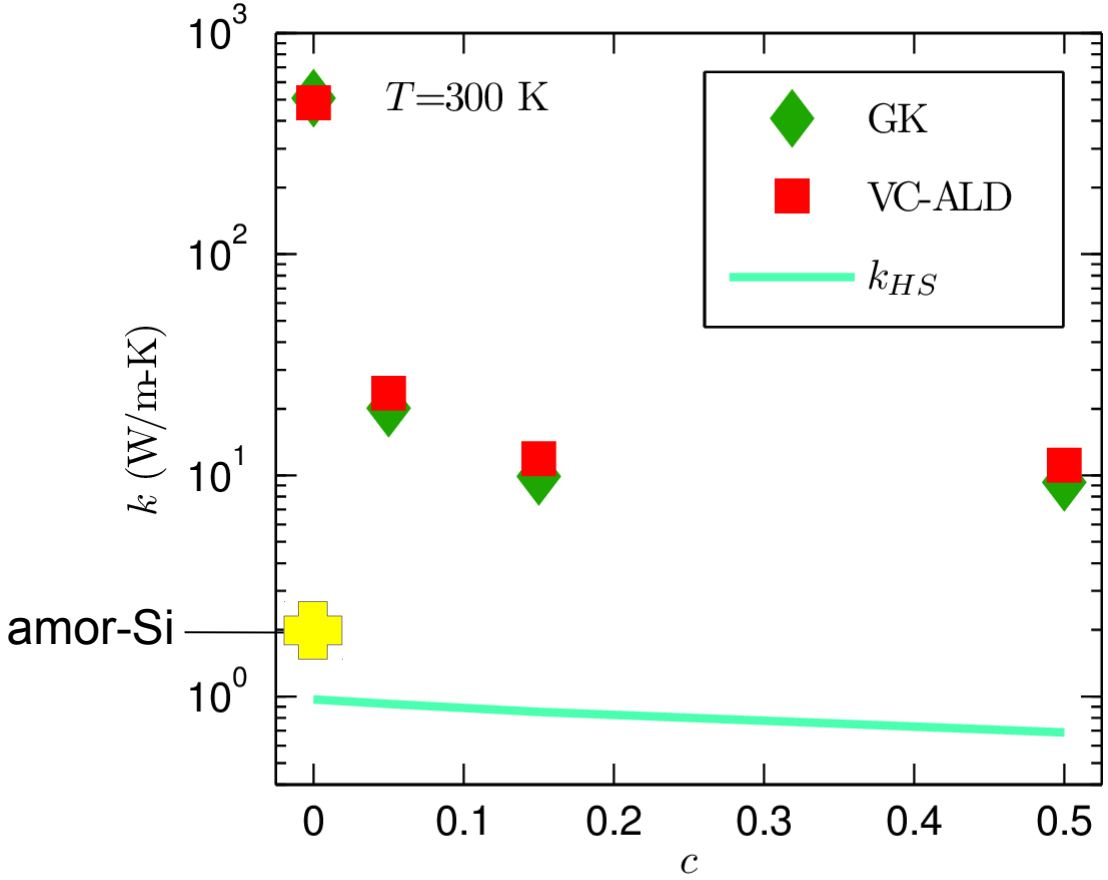




P. Sheng and M. Zhou, Science 253, 539542 (1991)

Thermal conductivity: SW silicon alloy

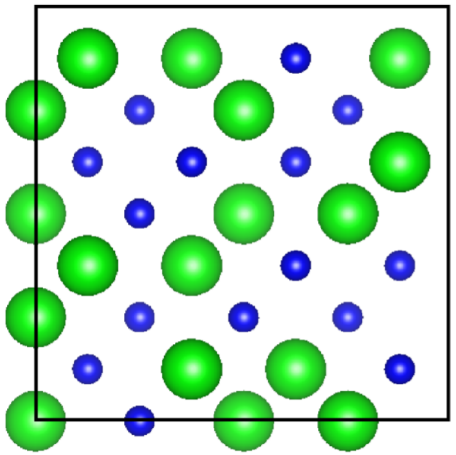


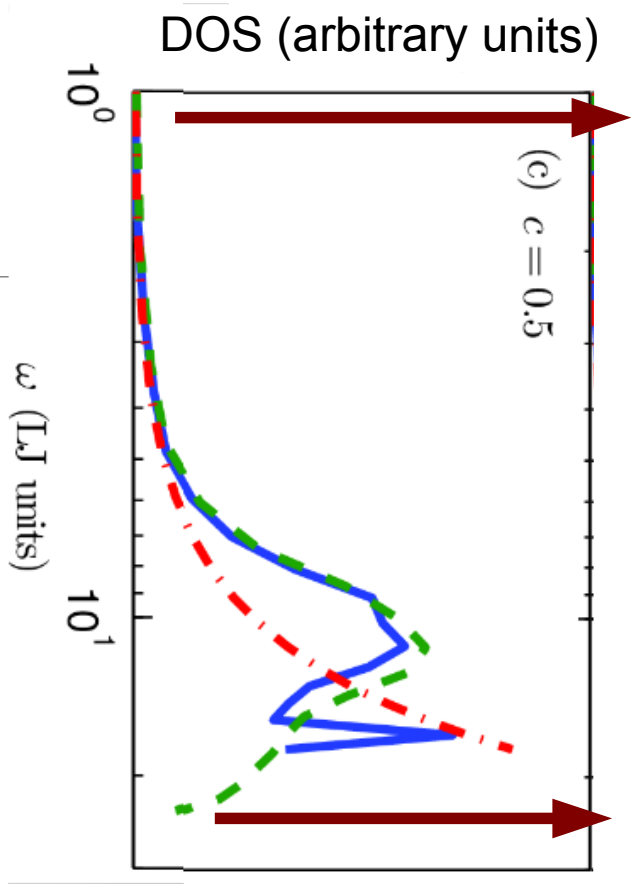


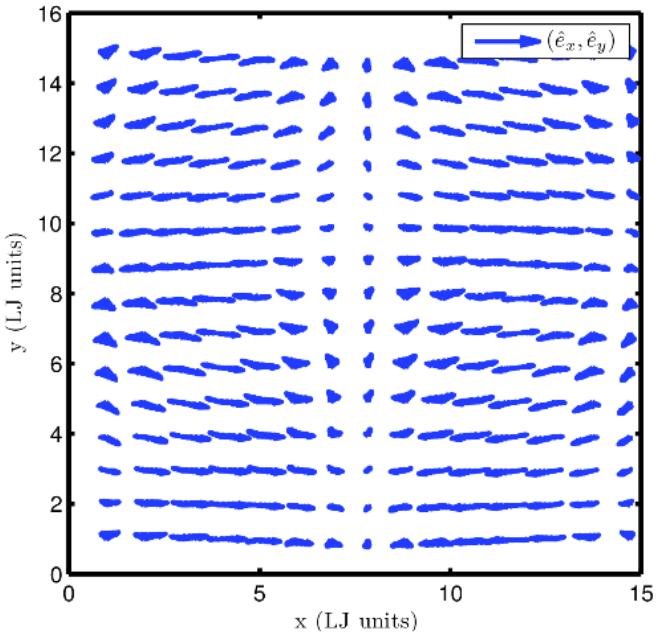
Gamma modes

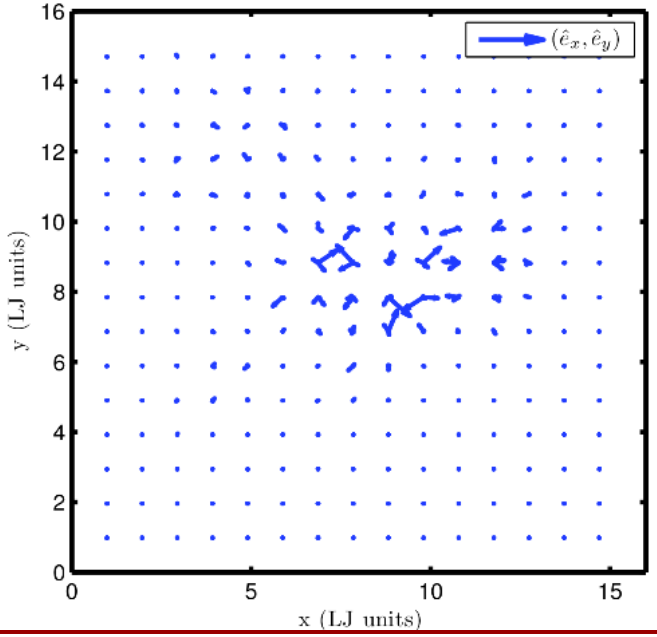
$e\left(\begin{smallmatrix}\kappa=0 & b\\ \nu & \alpha\end{smallmatrix}\right)$

(a) disordered supercell

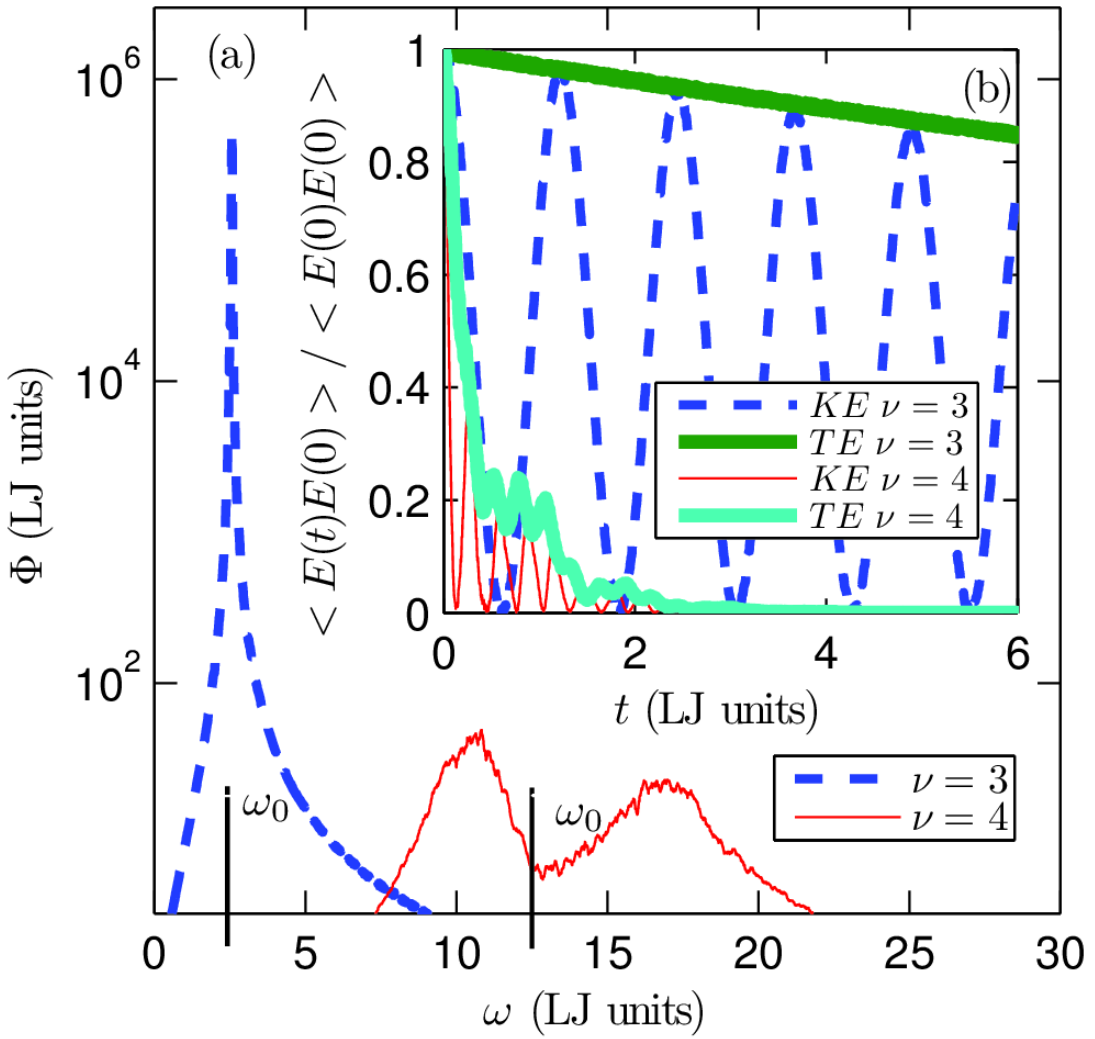




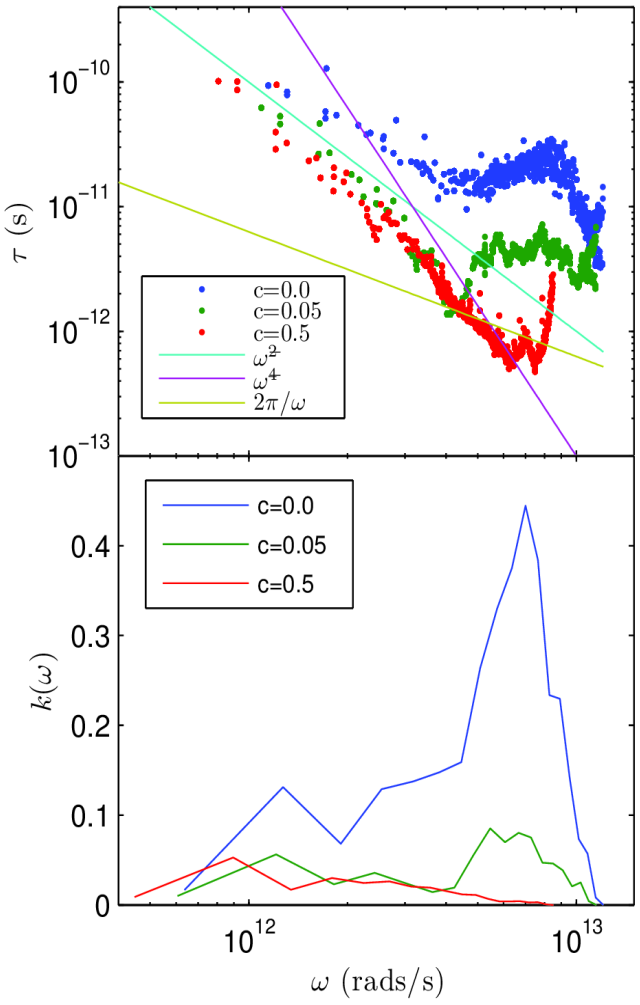


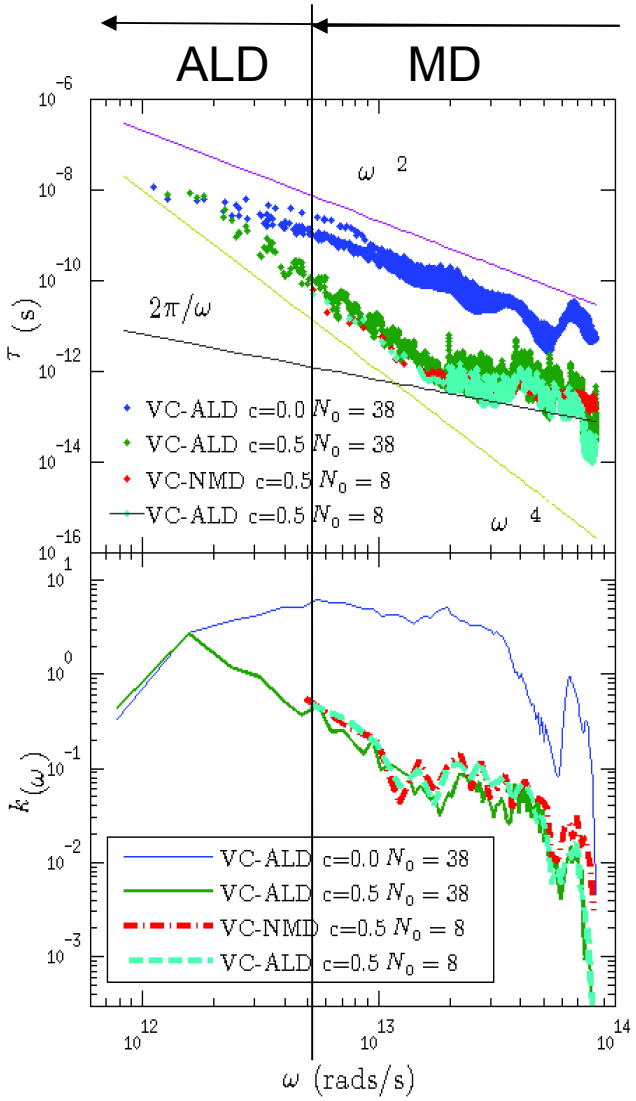


NMD using VC modes

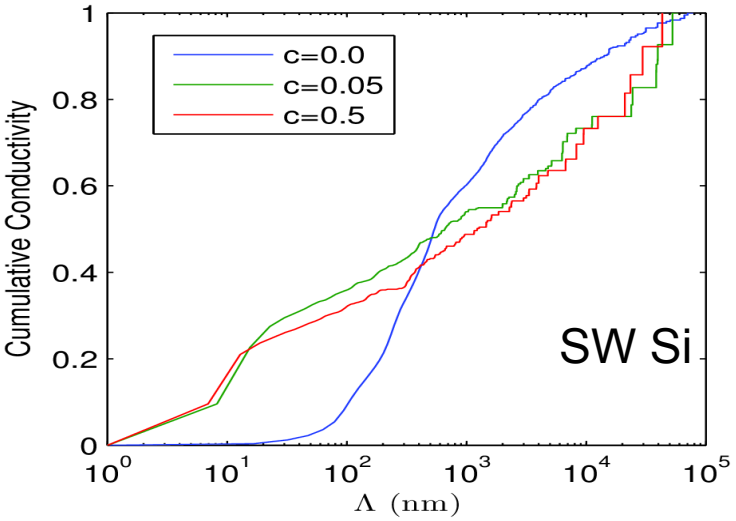


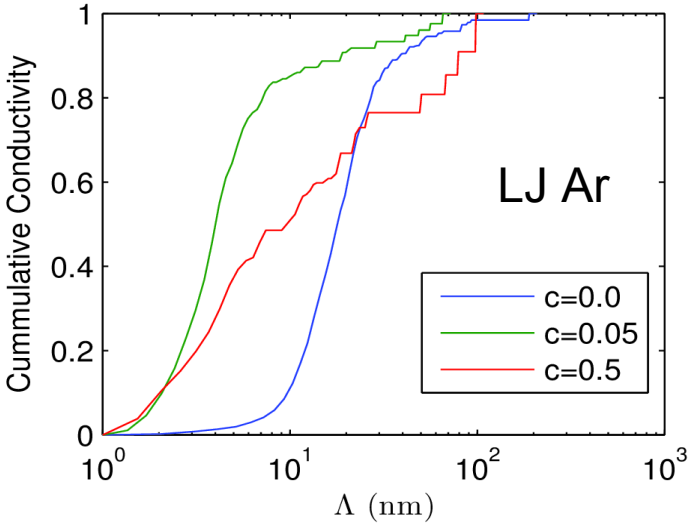
Phonon Spectrum: LJ Ar vs SW Si

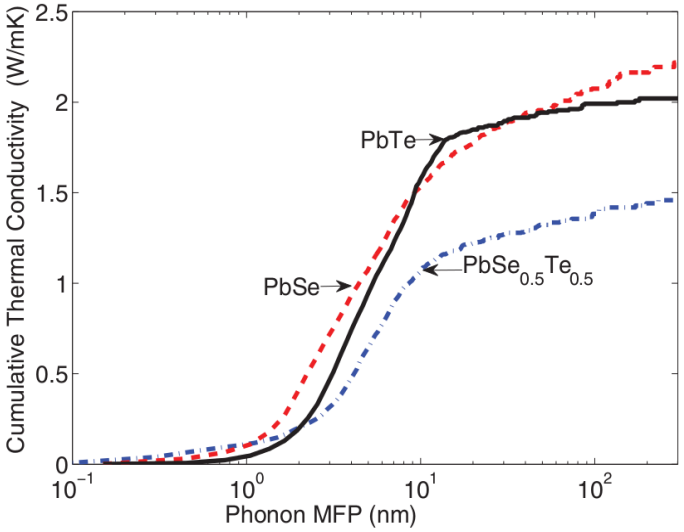




Conductivity Accumulation

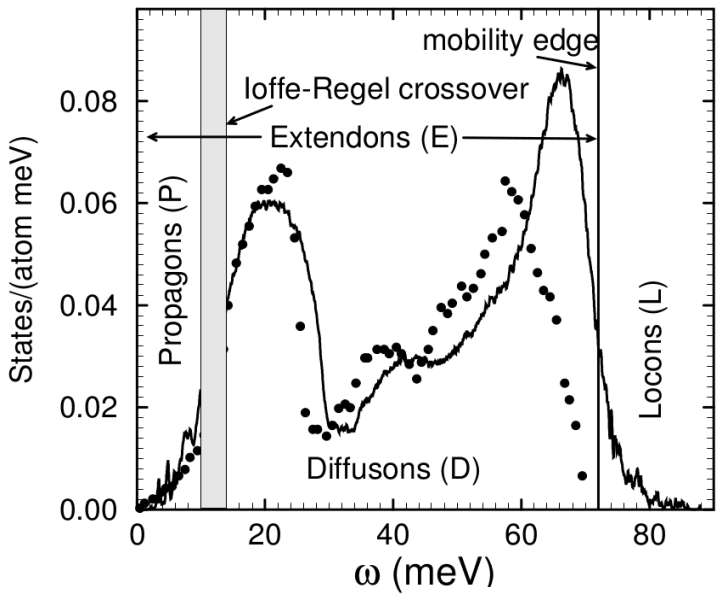


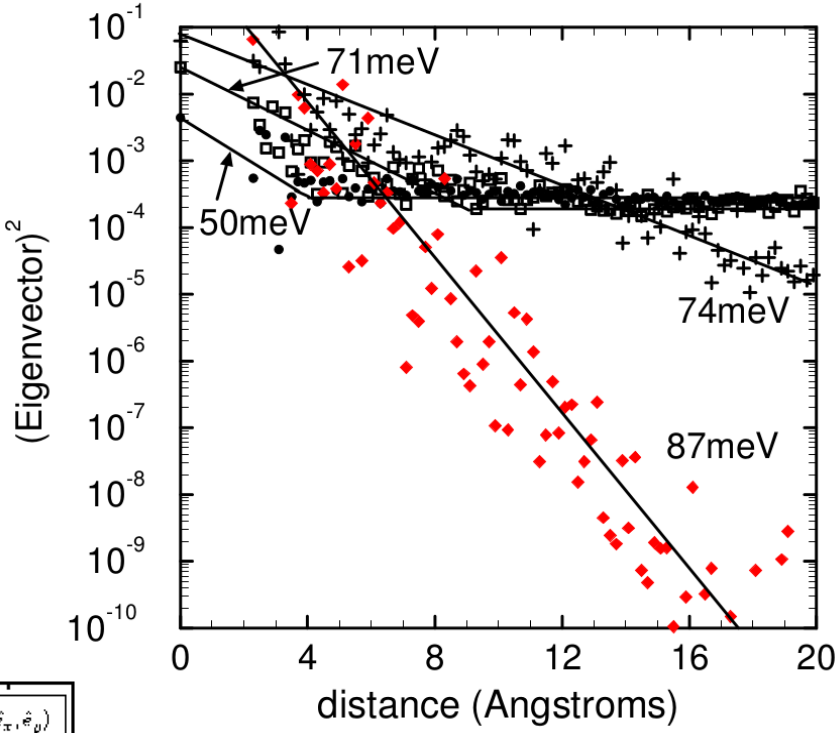


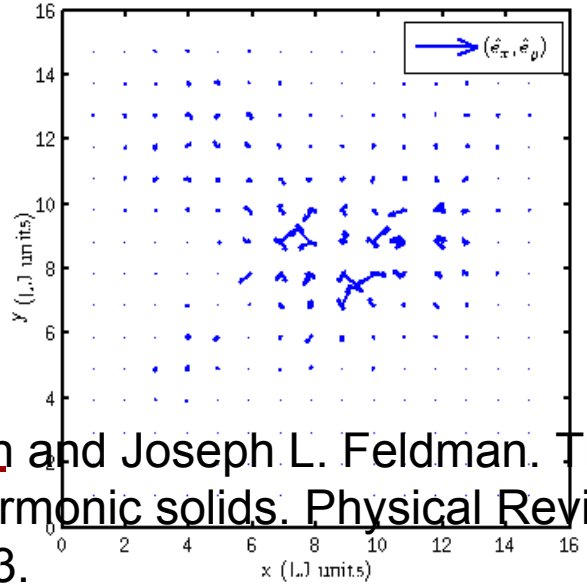


PHYSICAL REVIEW B 85, 184303 (2012)

propagons, diffusons, locons







[1] Philip B. Allen and Joseph L. Feldman. Thermal conductivity of disordered harmonic solids. Physical Review B, 48(17):12581–12588, Nov 1993.

Diamond

GaN

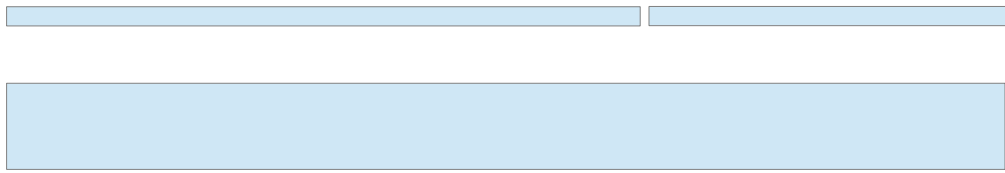
si

Si,HS

Si/Ge

PbTe,PbTe/Se,
(1/4)Tmelt = 300K

LJ,20K,
(1/4)T_melt



Exponential trends in Information Technologies

Moore's Law: 2^{n}

http://boards.straightdope.com/sdmb/showthread.php?t=316530

Human Genome

http://en.wikipedia.org/wiki/Kurzweil_Music_Systems

Exponential trends music: orchestra

1980: $100,000

http://boards.straightdope.com/sdmb/showthread.php?t=316530

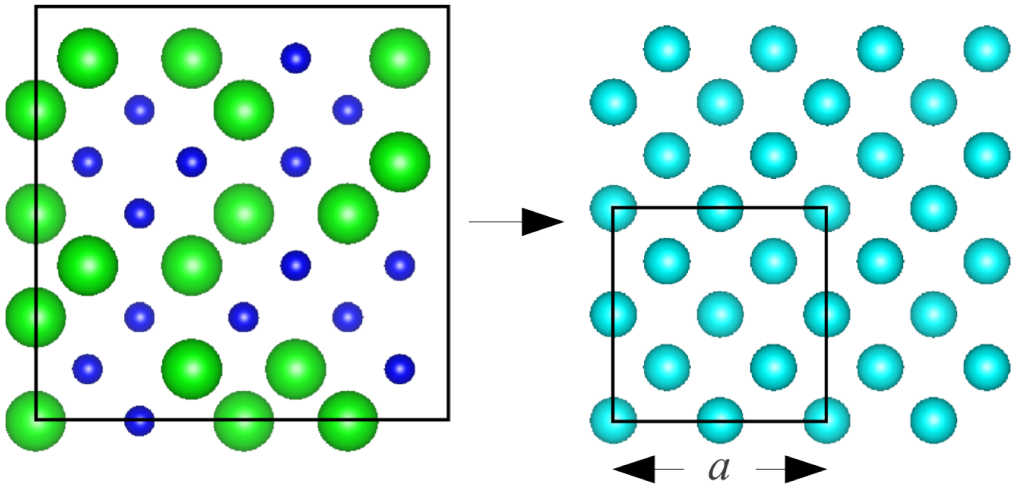
2003: $2,000 (my setup

http://en.wikipedia.org/wiki/Kurzweil_Music_Systems

VC-ALD: Group Velocity

(a) disordered supercell

(b) VC unit cell



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$$D_{ph}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right) \approx 0$$