Evaluation of the Virtual Crystal Approximation for Predicting Thermal Conductivity

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04/04/13

Carnegie Mellon

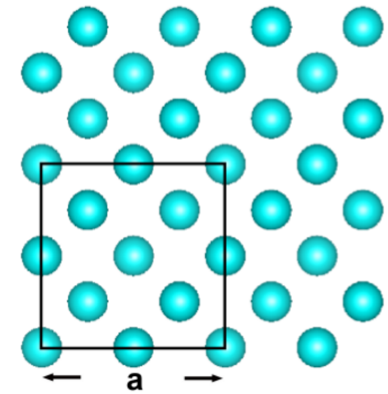
Mechanical Engineering

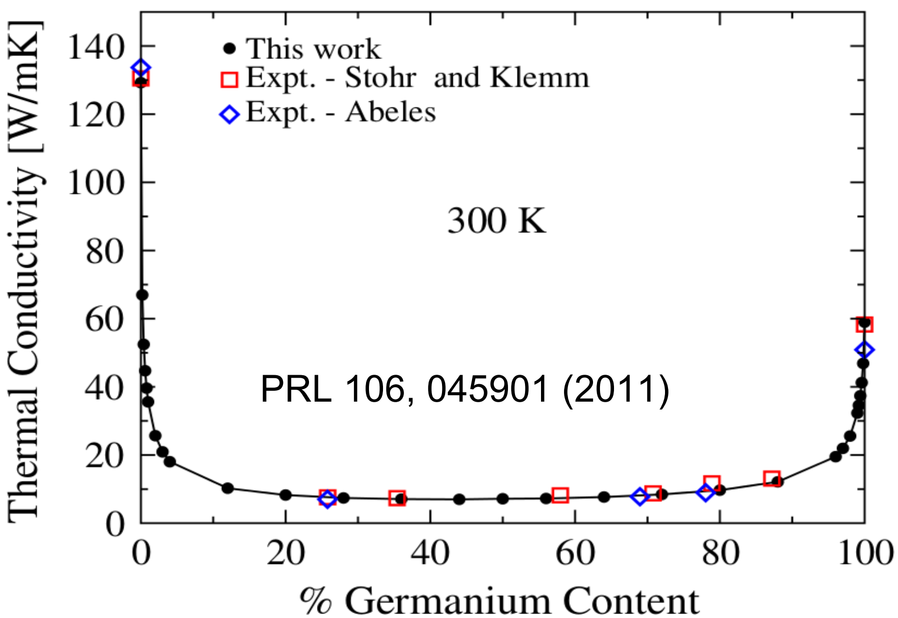
Motivation: experimental accuracy

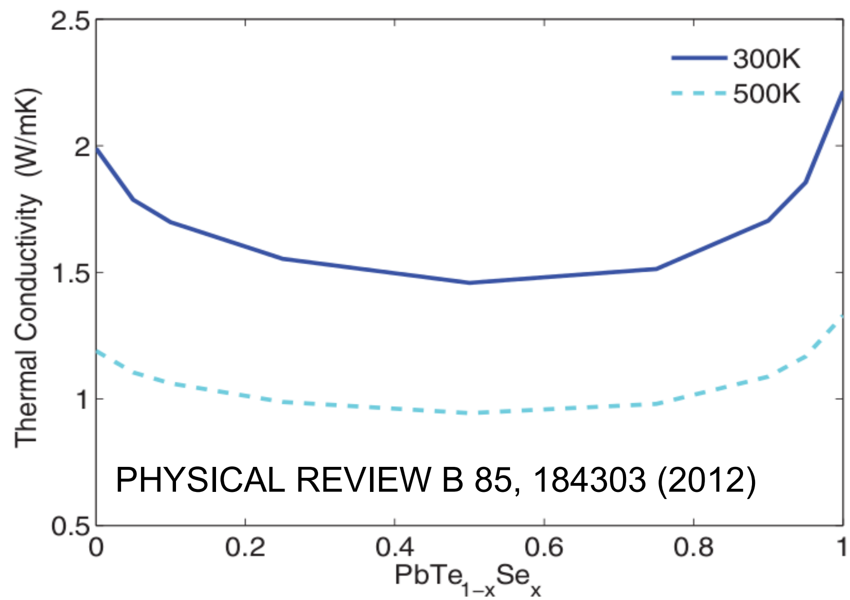
Expensive Density Functional Theory (DFT): force constants

Anharmonic Lattice Dynamics (ALD): based on unit cell

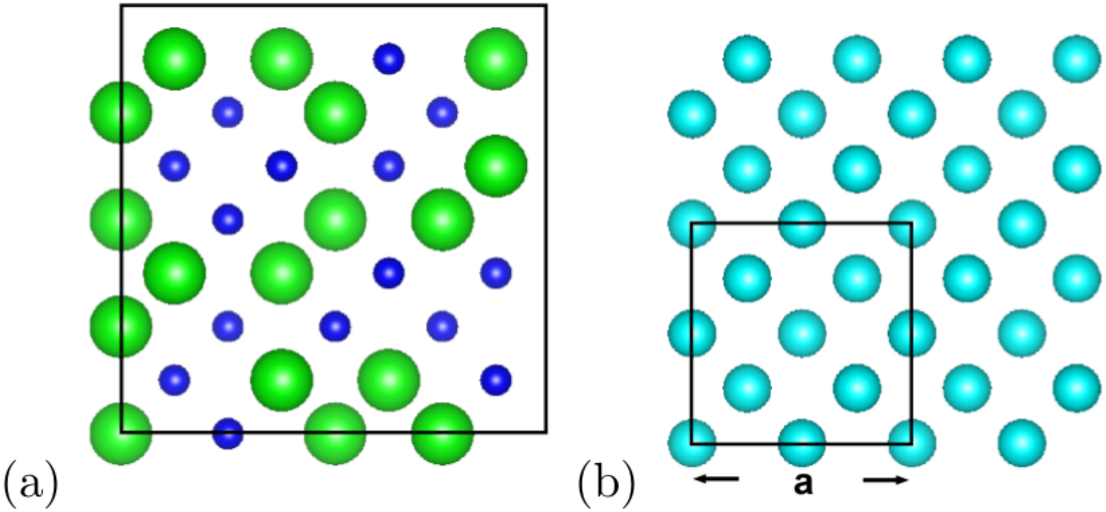
Alloys: isotopic effects, thermoelectric materials







Virtual Crystal Approximation



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

disorder strength:

$$g_n = \sum_{\mu} c^{\mu} (1 - m^{\mu}/\bar{m}^{\mu})^n$$

Kinetic Theory for Crystal

single-mode relaxation time approximation + Boltzmann Transport Equation + classical harmonic limit:

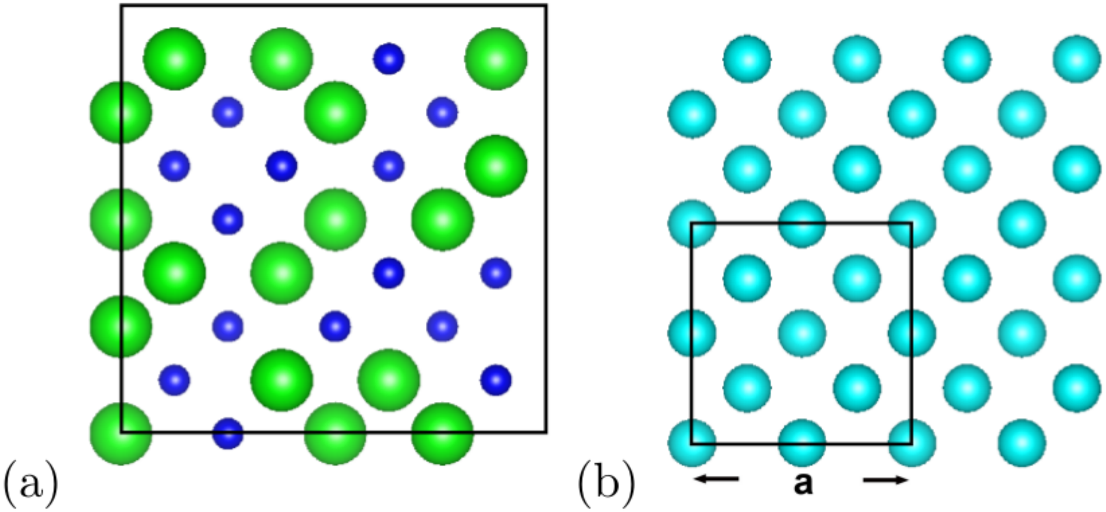
$$k_{ph,\mathbf{n}} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} \frac{k_B}{V} D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu}$$

mode thermal diffusivity:

$$D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = v_{g,\mathbf{n}}^2\binom{\boldsymbol{\kappa}}{\nu} \tau\binom{\boldsymbol{\kappa}}{\nu}$$

Macro-theory: k = \rho C_p \alpha

VC Approximation with ALD (VC-ALD)



$$D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = v_{g,\mathbf{n}}^{2}\binom{\boldsymbol{\kappa}}{\nu}\,\tau\binom{\boldsymbol{\kappa}}{\nu}$$

ALD

$\tau_{p-p} \sim 1/\,\omega^{2}$

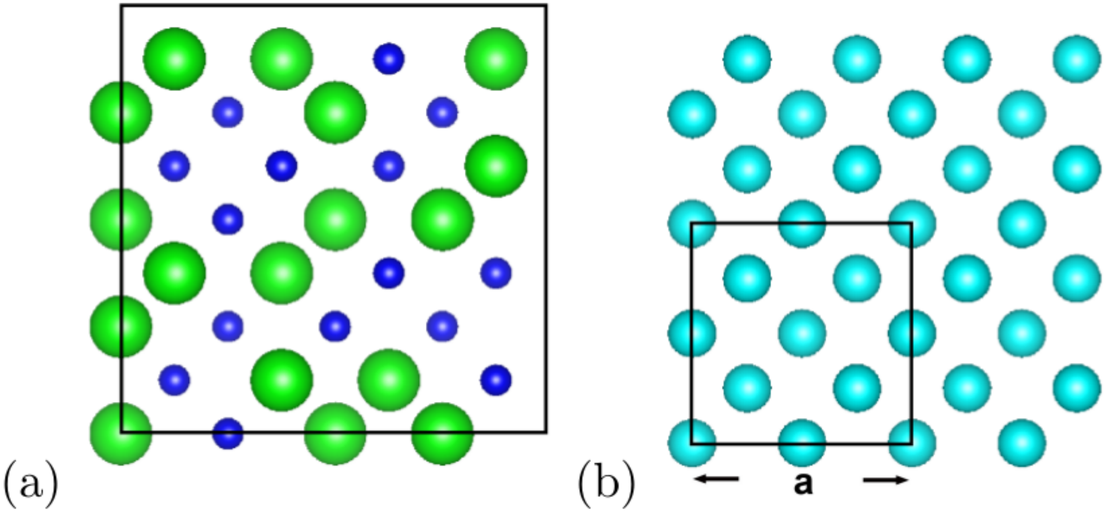
Tamura

$\tau_{p-d}\binom{\boldsymbol{\kappa}}{\nu} \sim 1/\,\omega^{4}$

Matthiessen's Rule

$$\frac{1}{\tau\binom{\boldsymbol{\kappa}}{\nu}} = \frac{1}{\tau_{p-p}\binom{\boldsymbol{\kappa}}{\nu}} + \frac{1}{\tau_{p-d}\binom{\boldsymbol{\kappa}}{\nu}}$$

VC Approximation with ALD (VC-ALD)

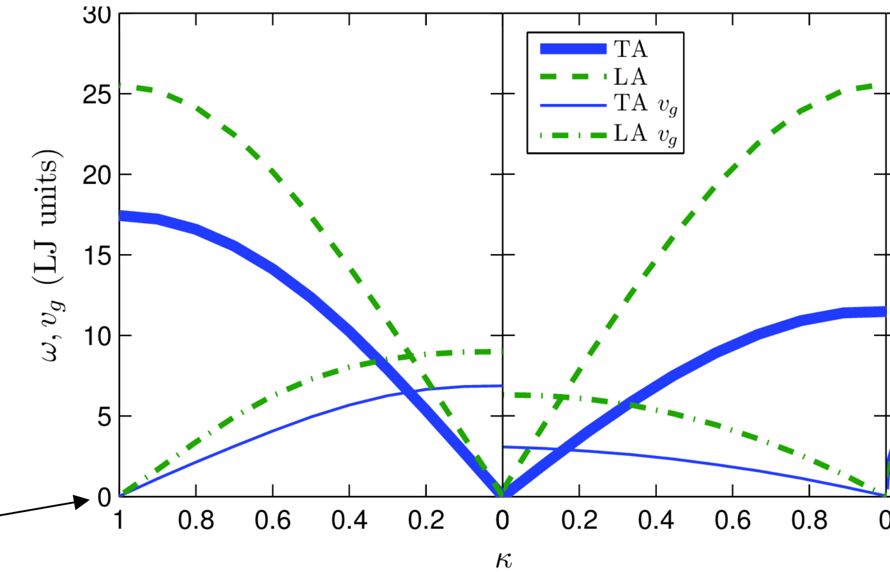


$$D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = v_{g,\mathbf{n}}^{2}\binom{\boldsymbol{\kappa}}{\nu}\,\tau\binom{\boldsymbol{\kappa}}{\nu}$$

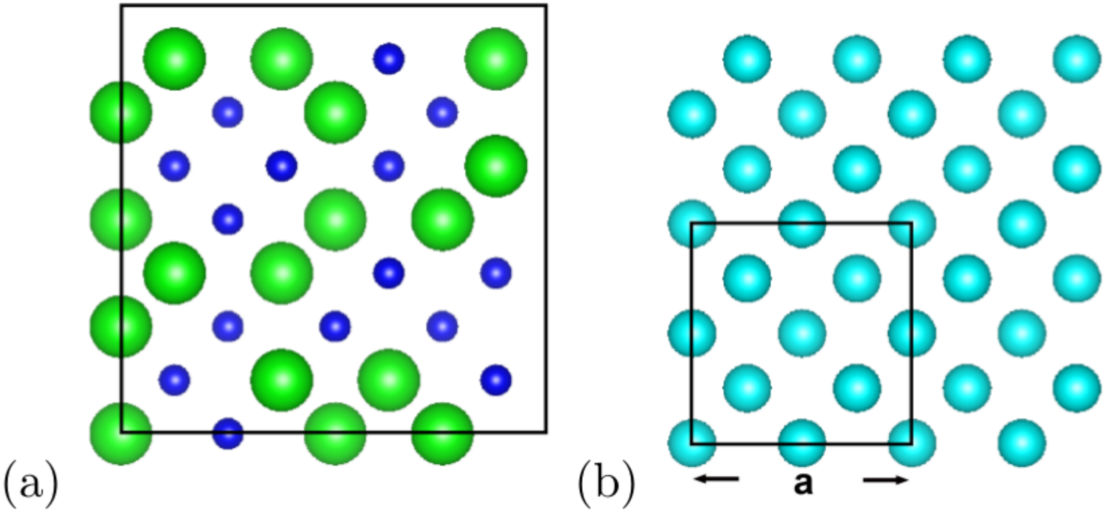
$$v_{g,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = \partial\omega\binom{\boldsymbol{\kappa}}{\nu}\big/\partial\boldsymbol{\kappa}$$

$$\frac{\partial\omega\binom{\boldsymbol{\kappa}}{\nu}}{\partial\boldsymbol{\kappa}}$$

$$D_{ph}\binom{\boldsymbol{\kappa}}{\nu} \approx 0$$



Explicit disorder: Empirical Potentials

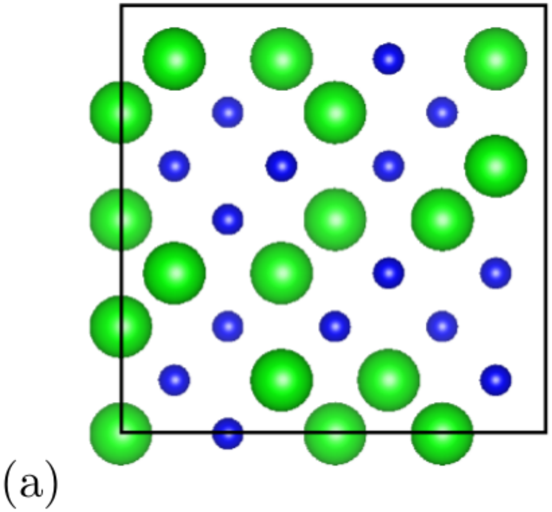


Virtual Crystal frequencies:

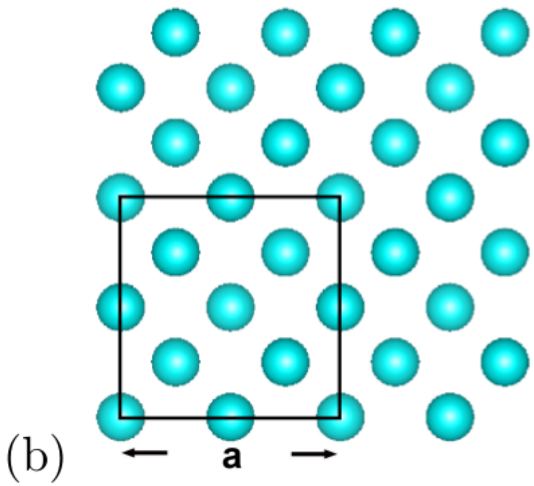
Lennard-Jones argon

Stillinger-Weber silicon

Explicit disorder: VC vs Gamma



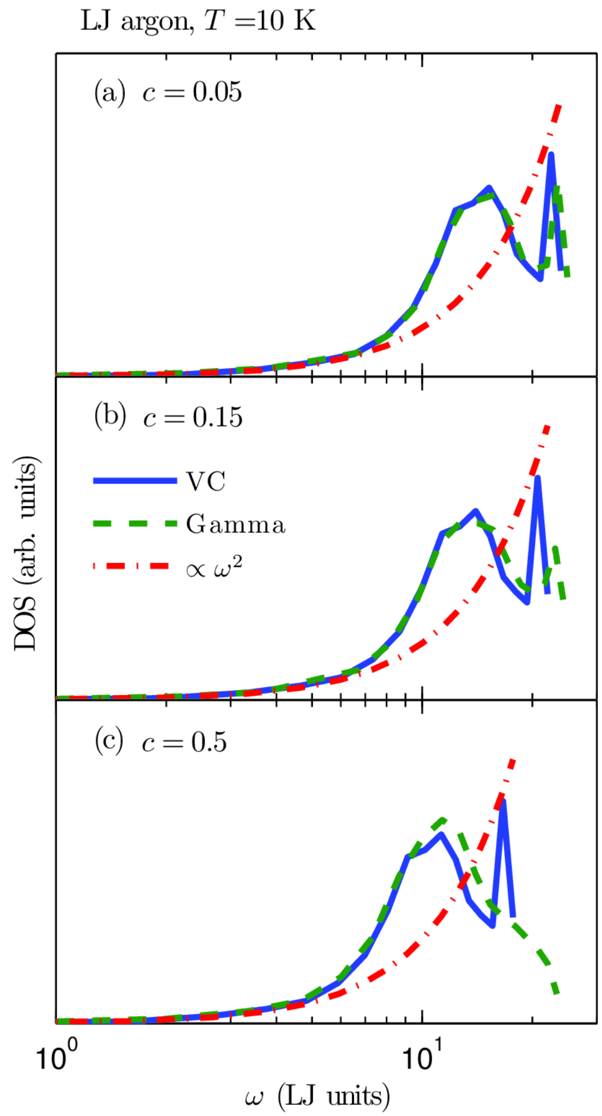
Gamma



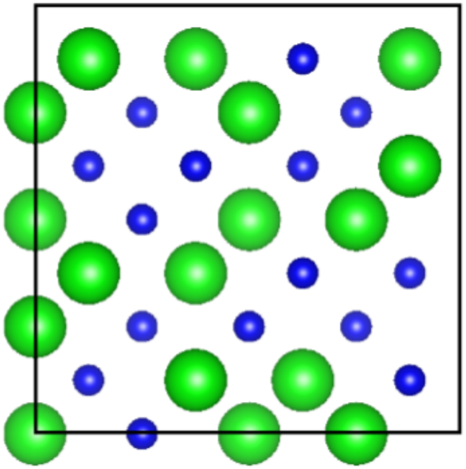
VC

Virtual Crystal frequencies:

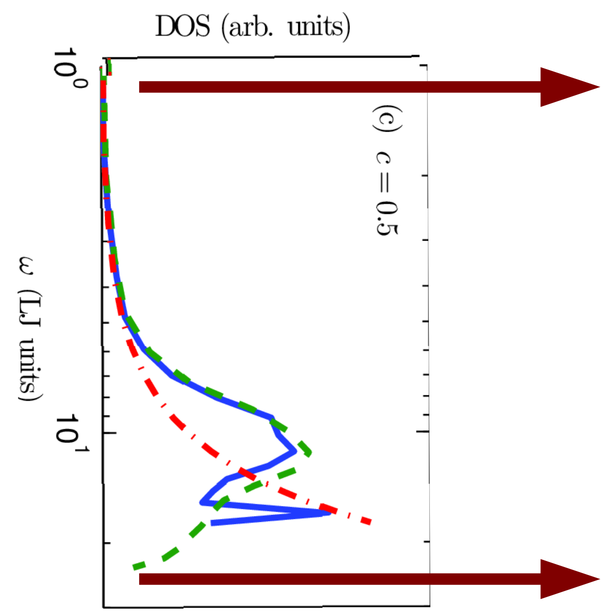
$$\omega \propto 1/[(1-c)m^i + cm^j]^{1/2}$$



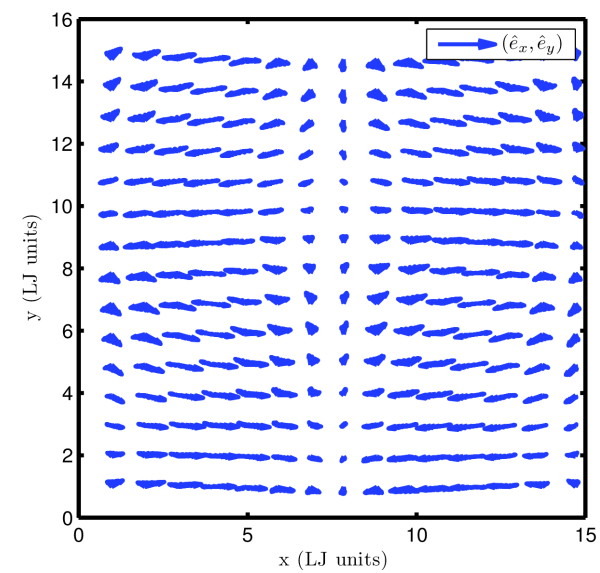
Gamma modes

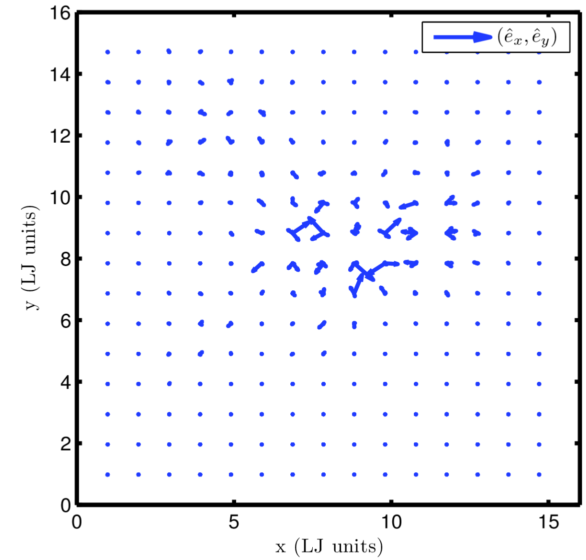


(a)



$e\begin{pmatrix}\boldsymbol{\kappa}=0 & b \\ \nu & \alpha\end{pmatrix}$

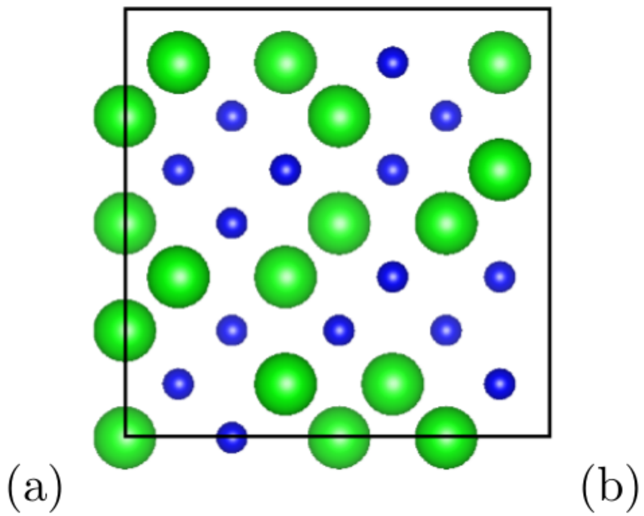


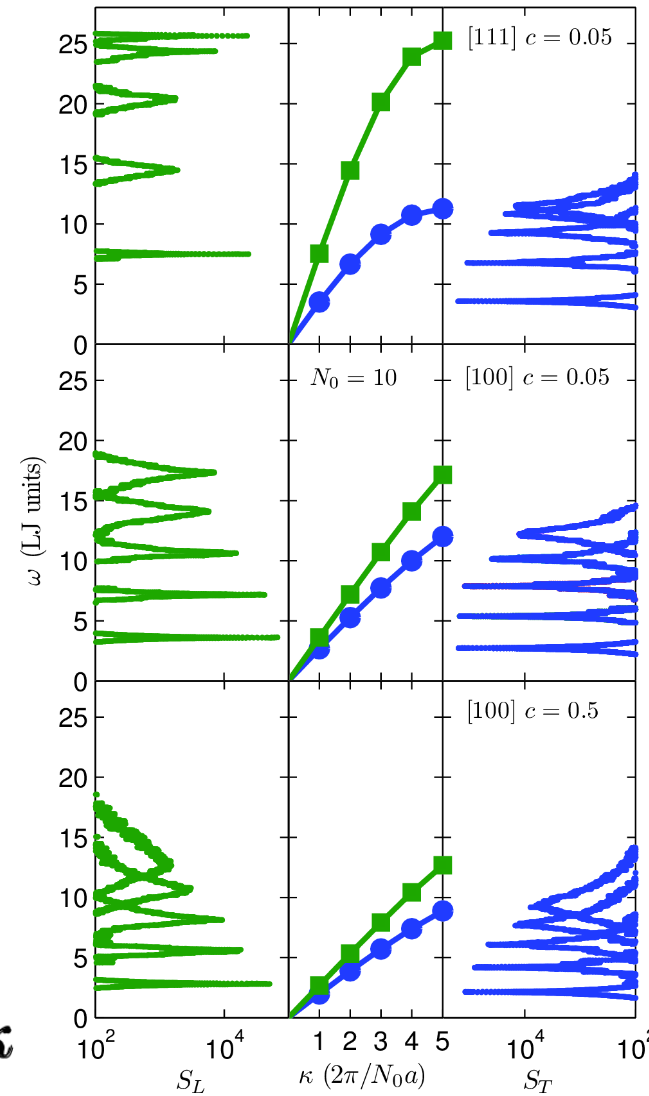


longitudinal (and transverse) polarizations:

$$E^L\begin{pmatrix}\boldsymbol{\kappa}_{VC} \\ \nu\end{pmatrix} = \left| \sum_b \hat{\boldsymbol{\kappa}}_{VC} \cdot e\begin{pmatrix}\boldsymbol{\kappa}=0 & b \\ \nu & \alpha\end{pmatrix} \exp\left[i\boldsymbol{\kappa}_{VC} \cdot \boldsymbol{r}_0\begin{pmatrix} l=0 \\ b\end{pmatrix}\right] \right|^2$$

Gamma modes Structure Factor





longitudinal (and transverse) polarizations:

VC group velocity:

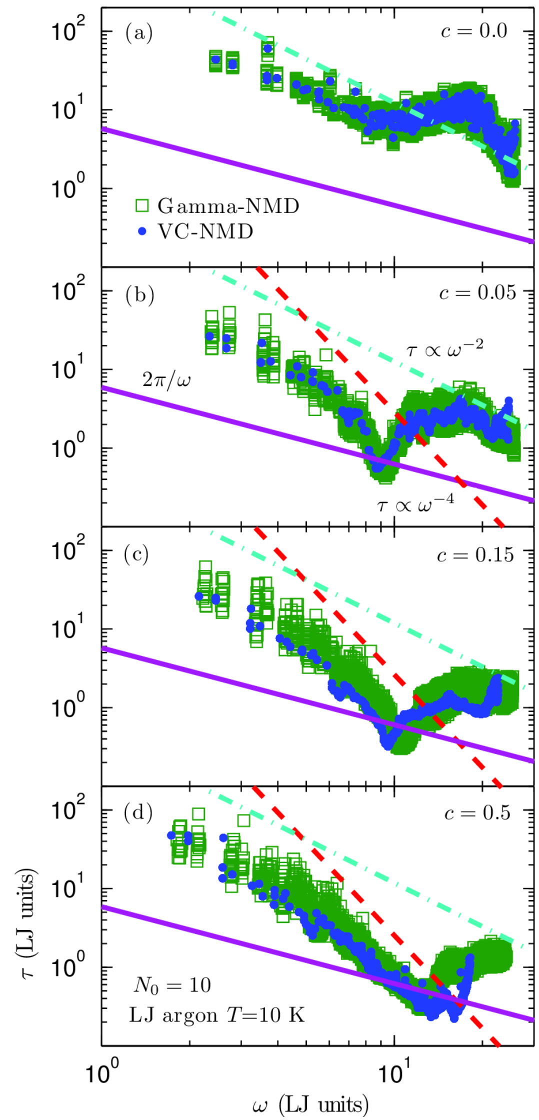
$$D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = v^2_{g,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu}\,\tau\binom{\boldsymbol{\kappa}}{\nu}$$

$$v_{g,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = \partial\omega\binom{\boldsymbol{\kappa}}{\nu}\Big/\partial\boldsymbol{\kappa}$$

Normal Mode Decomposition (NMD)

$$D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = v_{g,\mathbf{n}}^2\binom{\boldsymbol{\kappa}}{\nu}\,\tau\binom{\boldsymbol{\kappa}}{\nu}$$

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



Normal mode coordinate:

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

Normal mode energy:

$$E\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix};t\right) = \frac{\omega\binom{\boldsymbol{\kappa}}{\nu}^2}{2}q\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix};t\right)^*q\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix};t\right) + \frac{1}{2}\dot{q}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix};t\right)^*\dot{q}\left(\begin{smallmatrix}\boldsymbol{\kappa}\\ \nu\end{smallmatrix};t\right)$$

Allen-Feldman (AF), high-scatter limit

Phonons:

$$k_{ph,\mathbf{n}} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} \frac{k_B}{V} D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu}$$

Diffusons:

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

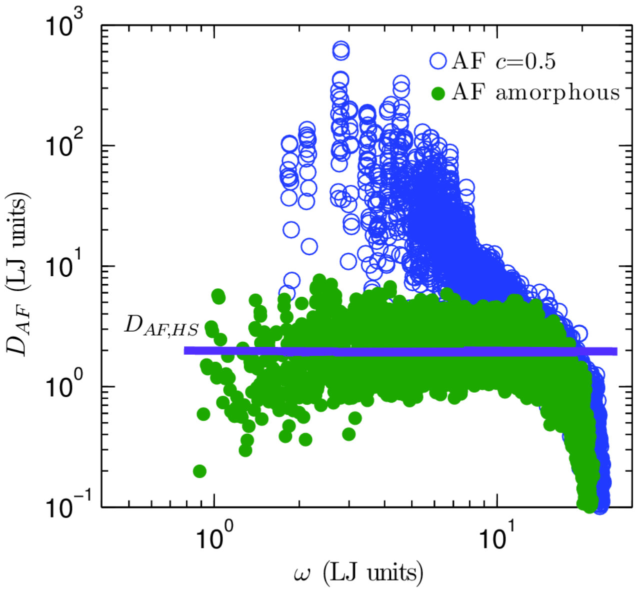
High-scatter limit:

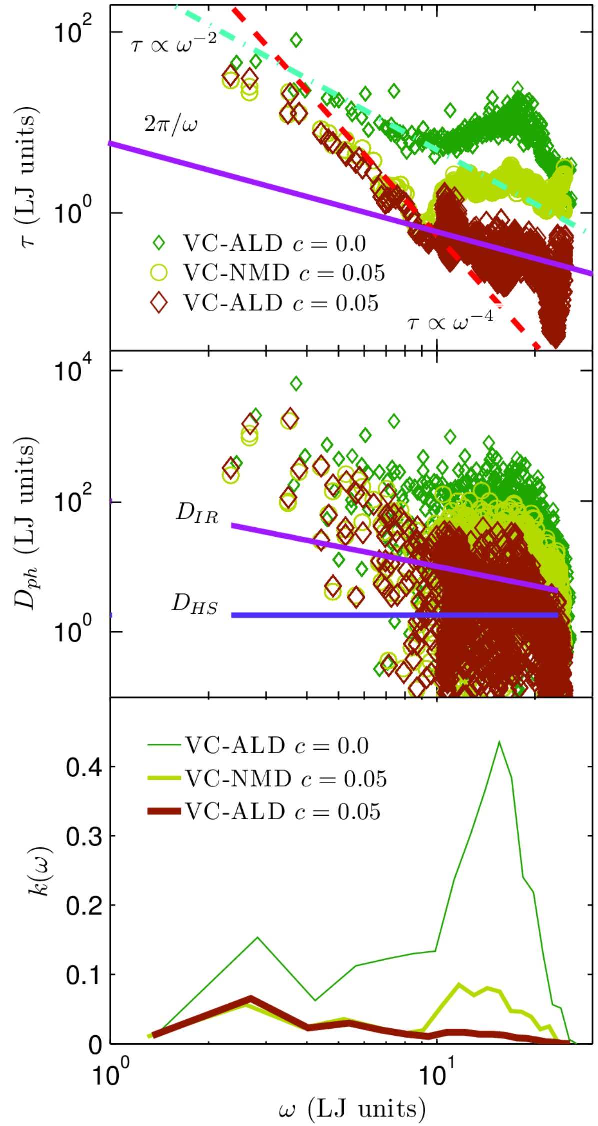
$$k_{HS} = \frac{k_B}{V_b} b v_s a \qquad D_{HS} = \frac{1}{3} v_s a$$

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

NMD, VC-ALD, AF thermal diffusivity

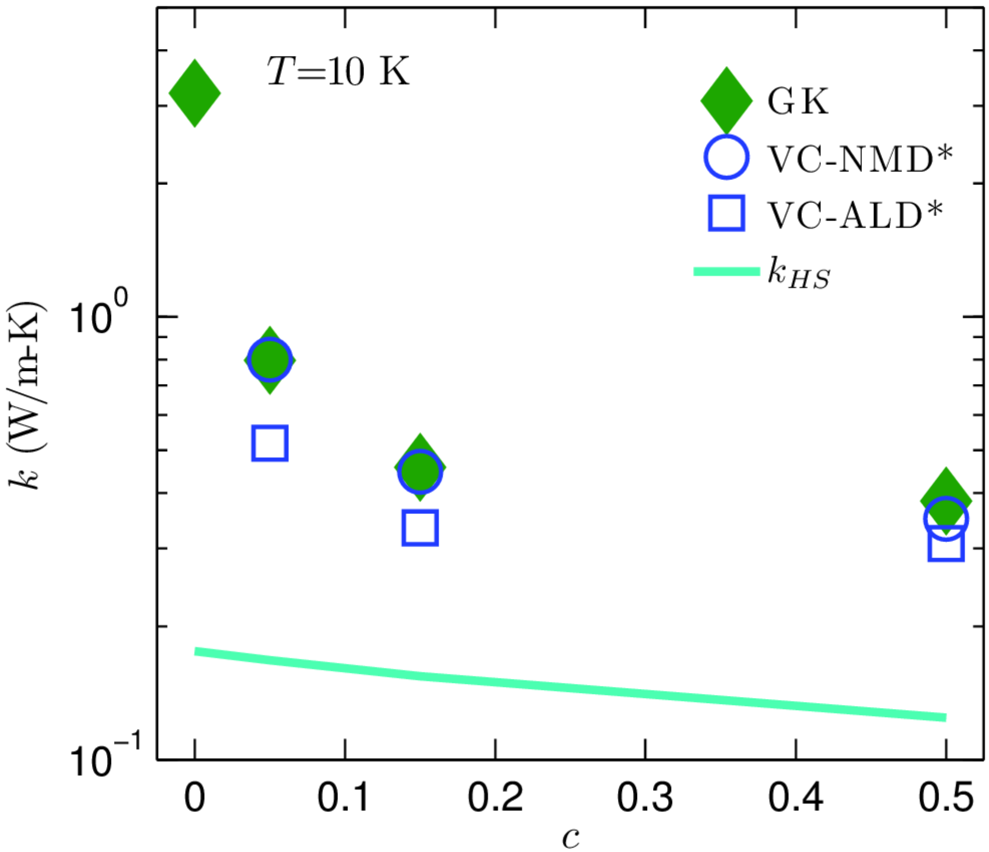
$$D_{ph}\binom{\boldsymbol{\kappa}}{\nu} < D_{HS}$$





Thermal conductivity: LJ argon alloy

Green-Kubo (GK): top-down method



High-scatter adjustment*:

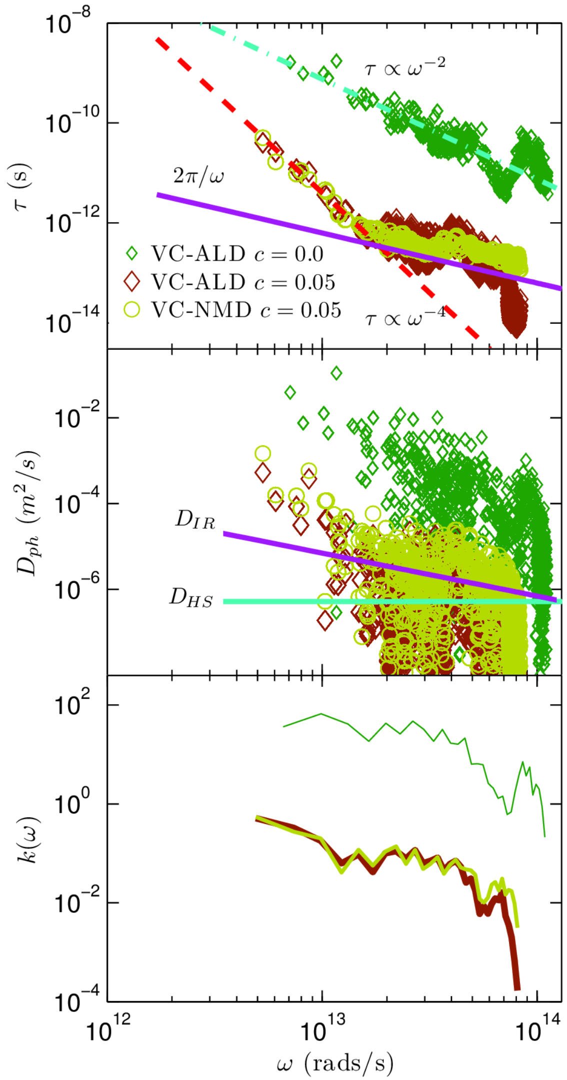
$$D_{ph}\binom{\boldsymbol{\kappa}}{\nu} < D_{HS}$$

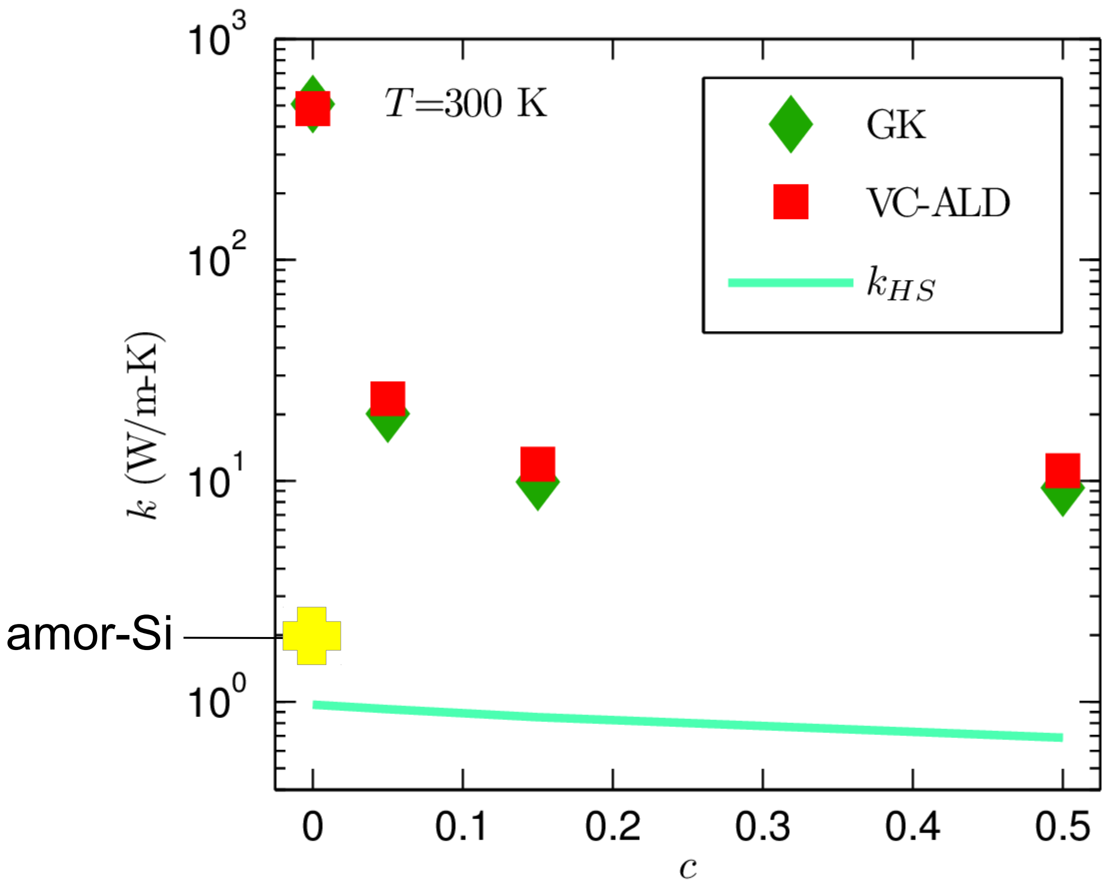
$$D_{ph}\binom{\boldsymbol{\kappa}}{\nu} = D_{HS}$$

amor-Ar

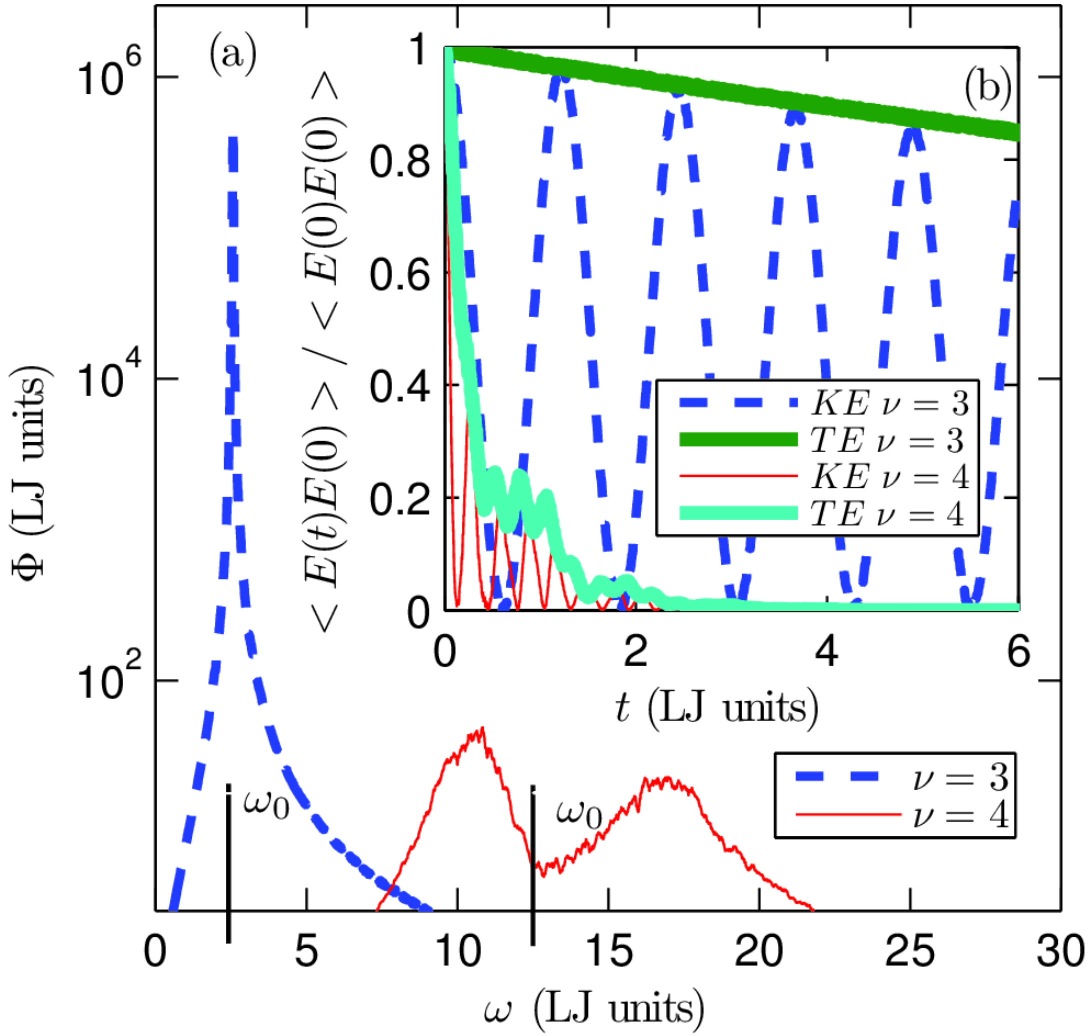
Summary

Thermal conductivity: SW silicon alloy



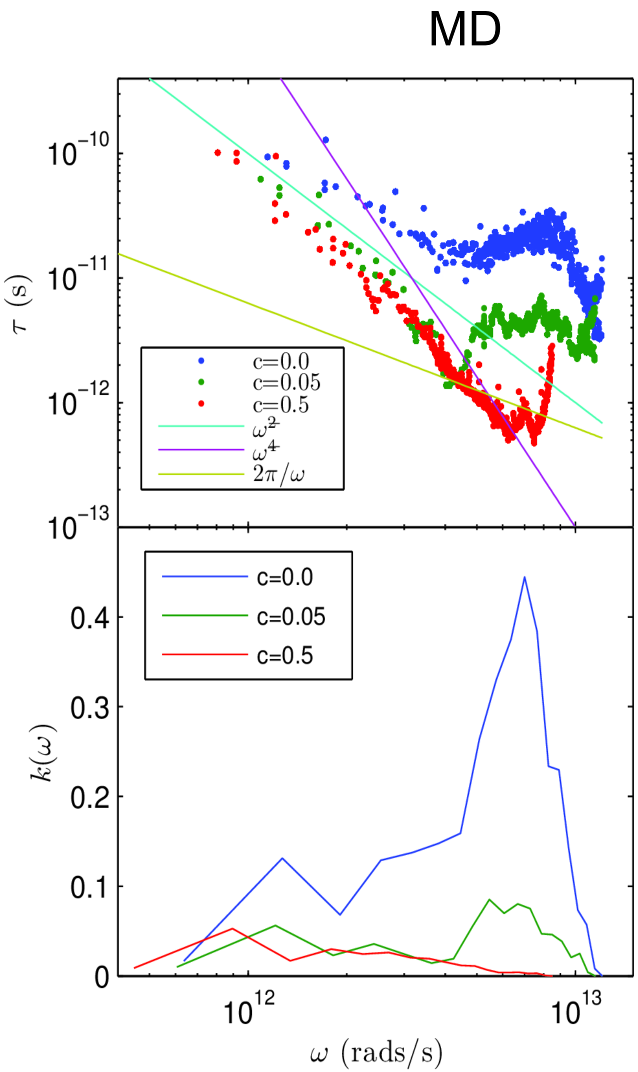


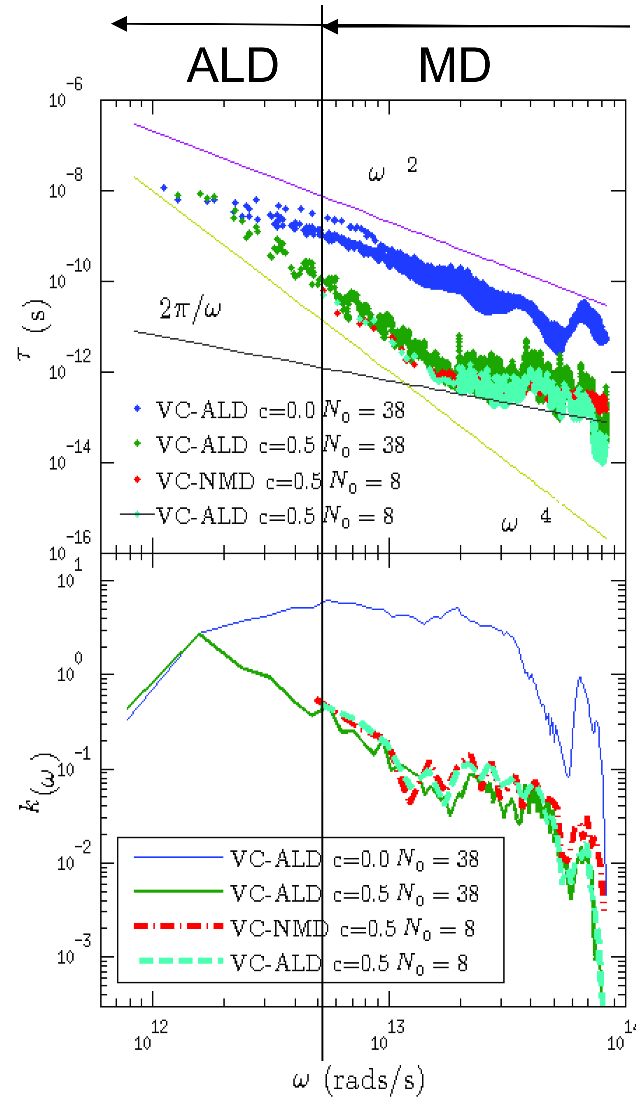
$$\frac{1}{\tau_{p-d}\binom{\boldsymbol{\kappa}}{\nu}} = \frac{\pi}{2} g_2 \omega^2\binom{\boldsymbol{\kappa}}{\nu}\, DOS\left(\omega\binom{\boldsymbol{\kappa}}{\nu}\right)$$



(a)
(b)
10^6
10^4
10^2
Φ (LJ units)
ω (LJ units)
$< E(t)E(0) > / < E(0)E(0) >$
t (LJ units)
ω_0
ω_0
$KE\ \nu = 3$
$TE\ \nu = 3$
$KE\ \nu = 4$
$TE\ \nu = 4$
$\nu = 3$
$\nu = 4$

Phonon Spectrum: LJ Ar vs SW Si





MD-based:

1E4 modes

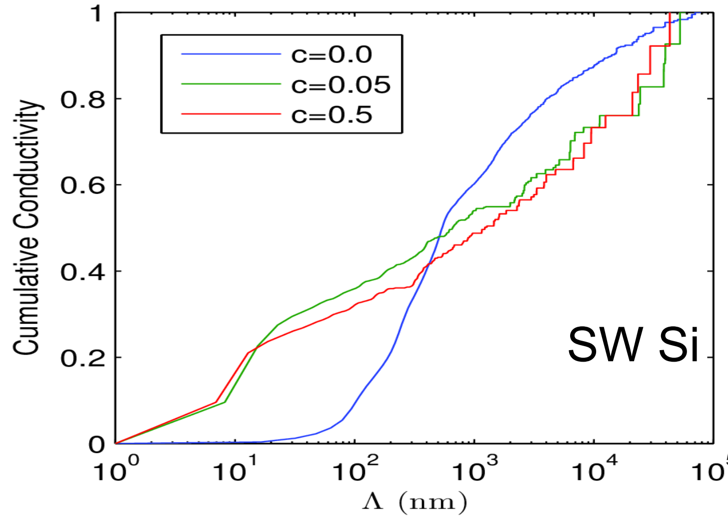
(7 days)*(100 cpu)

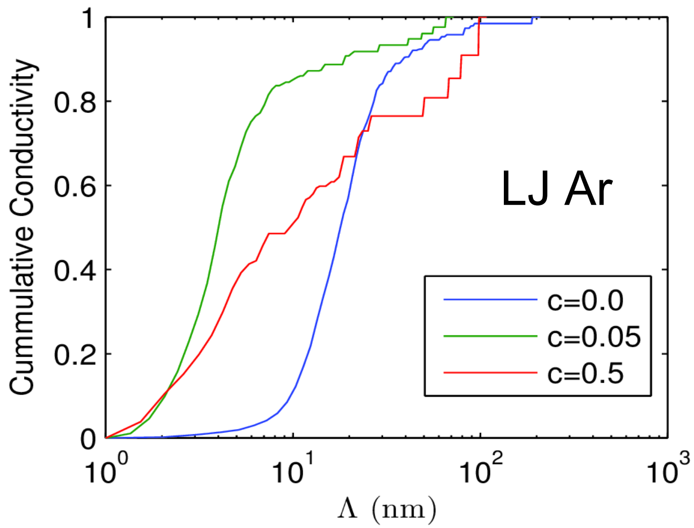
ALD:

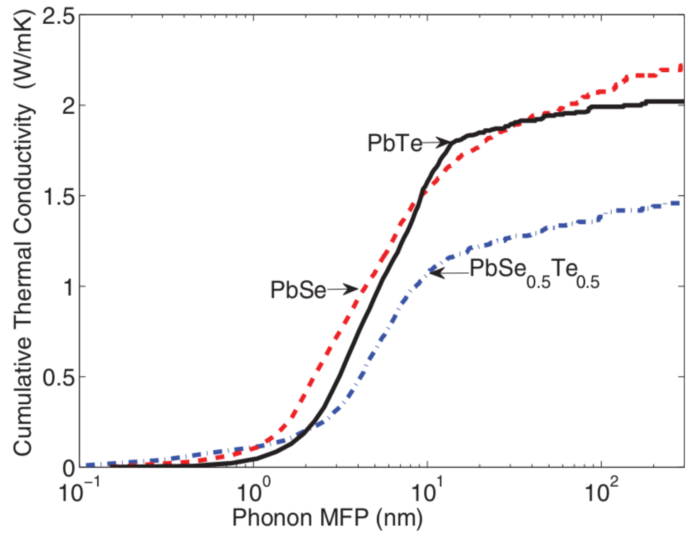
1E6 modes

(7 days)*(12 cpu)

Conductivity Accumulation







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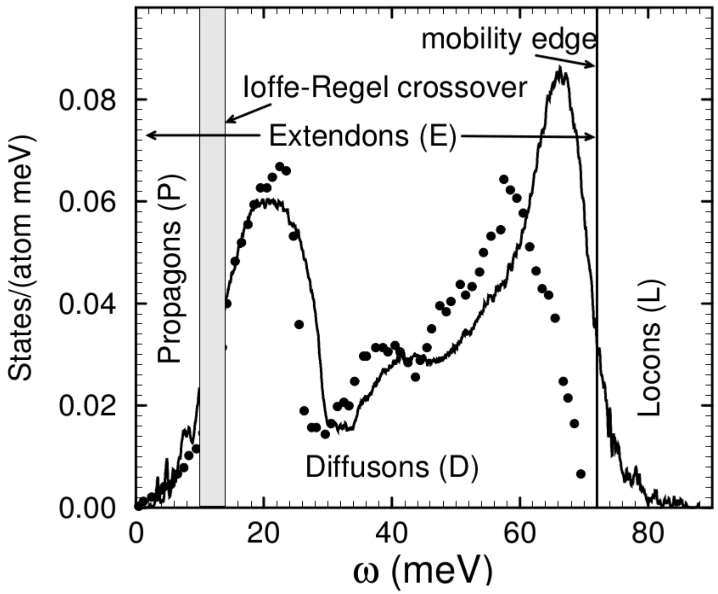
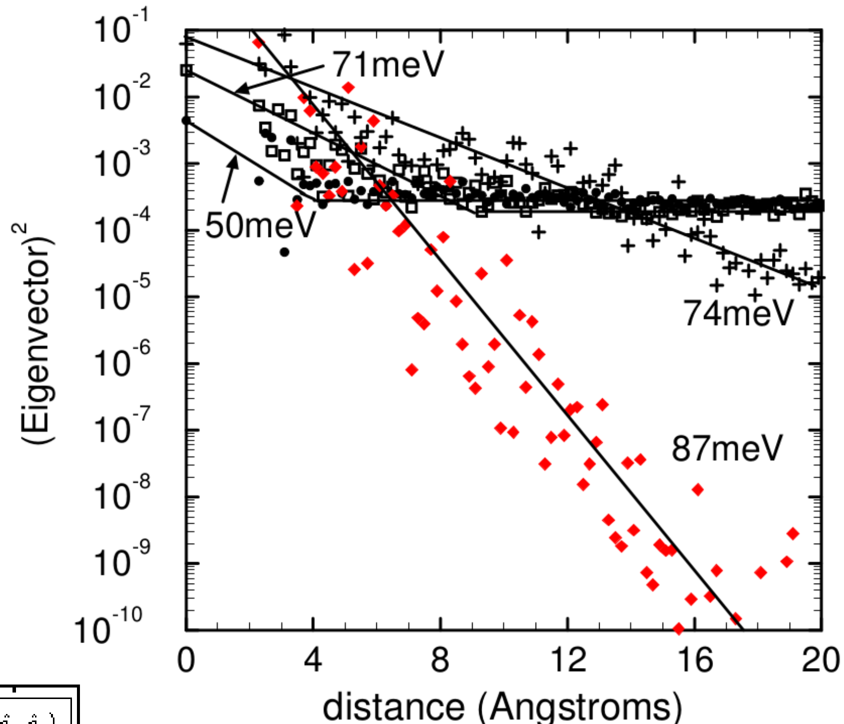
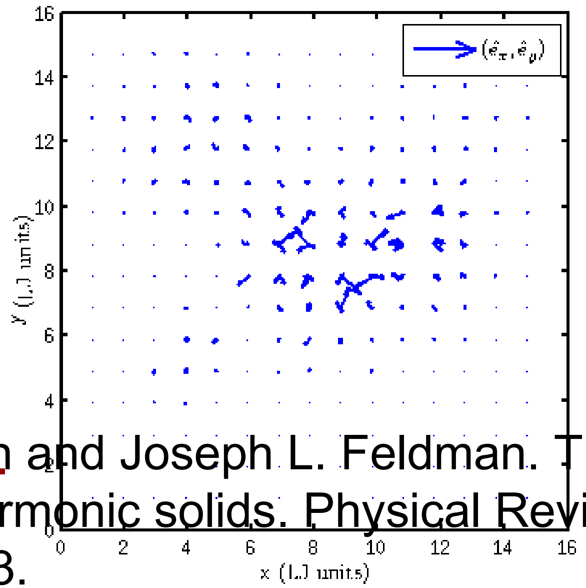
propagons, diffusons, locons







[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