Evaluation of the Virtual Crystal Approximation for Predicting Alloy Thermal Conductivity

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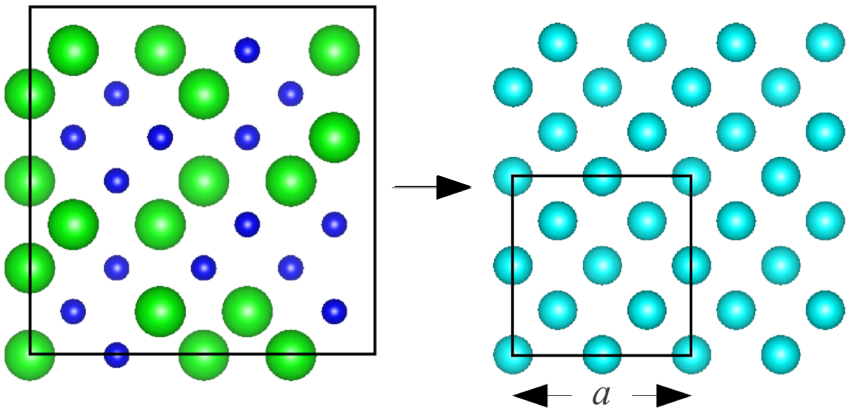
http://ntpl.me.cmu.edu/

04/04/13

Virtual Crystal Approximation

Gamma

VC



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

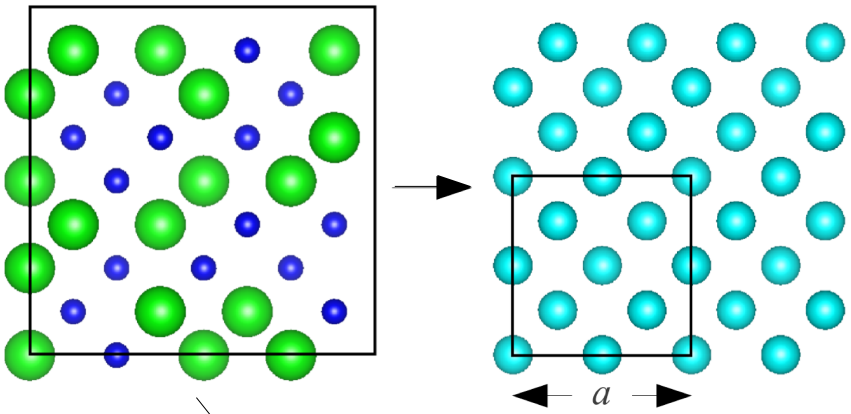
$$k_{ph,\mathbf{n}} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} \frac{k_B}{V} D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu}$$

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

Virtual Crystal: Diffusivities

Gamma

VC



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

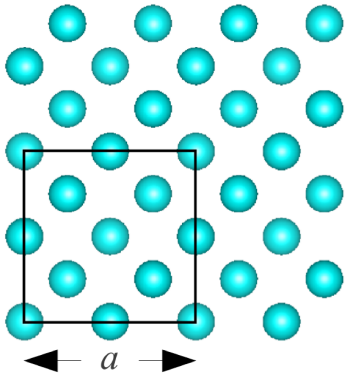
Allen-Feldman (**AF**) Theory:

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

$$D_{AF,i}(\omega_i) = v_g^2\,\tau \quad ?$$

VC-ALD Diffusivities: Lifetimes

VC



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

Perturbation theory:

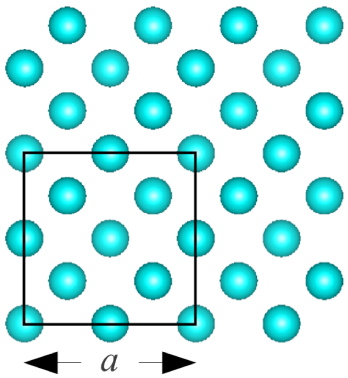
Anharmonic Lattice Dynamics (**ALD**)

Matthiessen's Rule:

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

VC-ALD Diffusivities: Lifetimes

VC



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

Perturbation theory:

Anharmonic Lattice Dynamics (**ALD**)

Phonon-Defect[1]

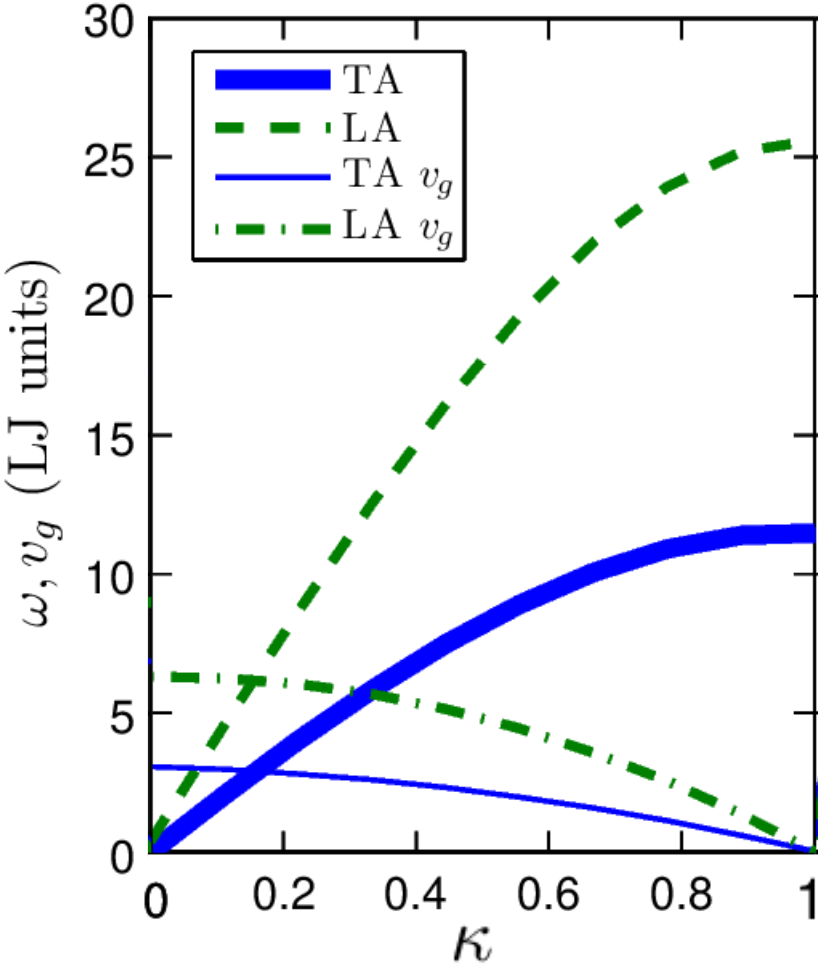
Matthiessen's Rule:

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

[1]Physical Review B 27, 858866 (1983)

VC-ALD Diffusivities: Group Velocity

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



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

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

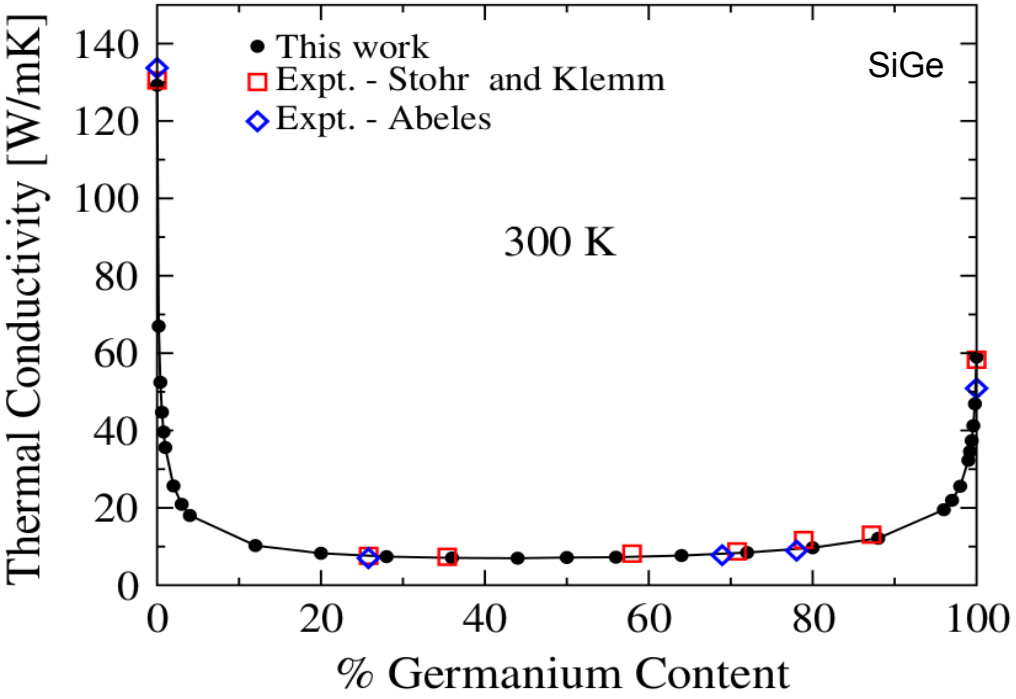
High-Scatter limit:

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

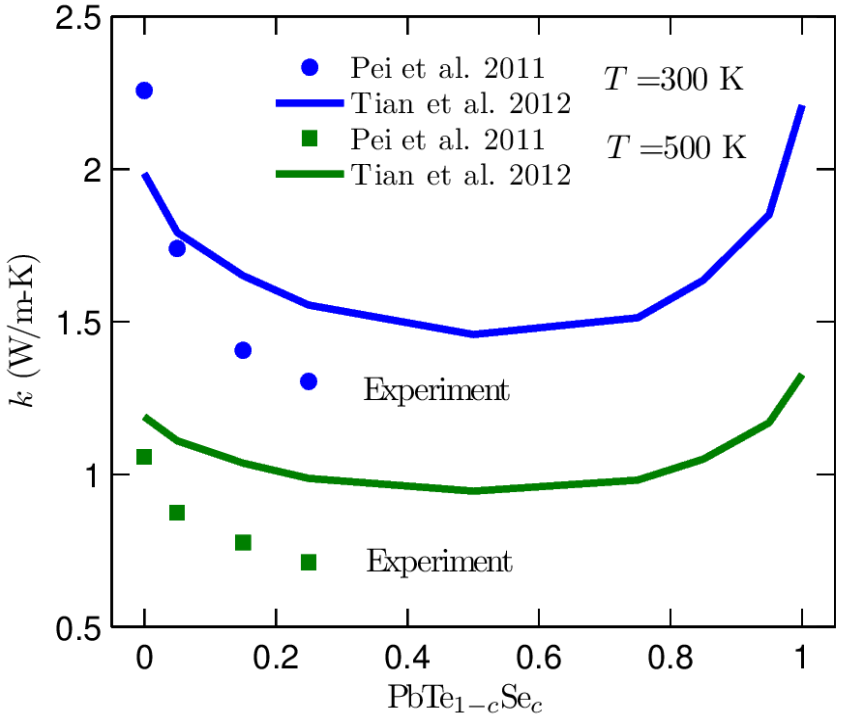
[1] Ann. Rev. of Phys. Chem. 39, 93121 (1988)

VC-ALD: experimental accuracy

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



PRL 106, 045901 (2011)

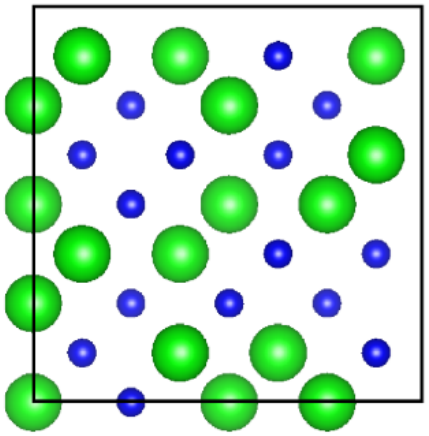


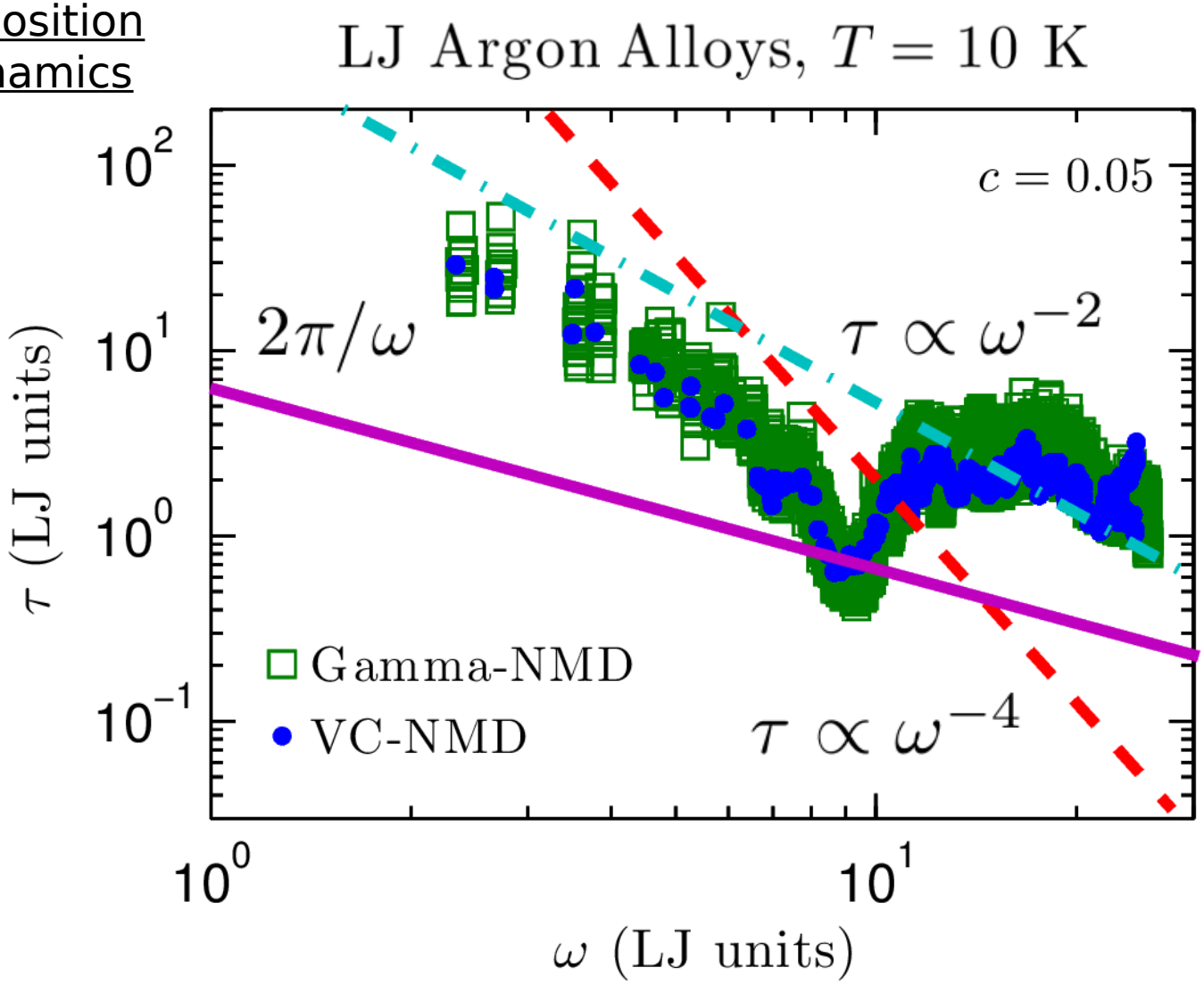
PRB 85, 184303 (2012)

Explicit disorder: NMD

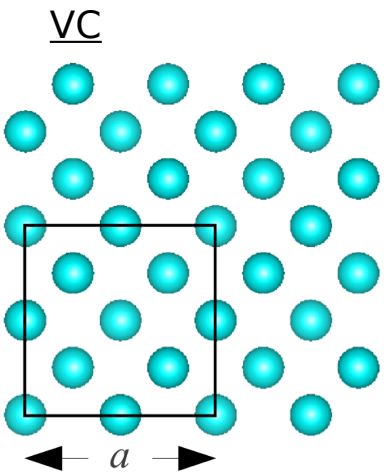
Normal Mode Decomposition (**NMD**): Molecular Dynamics

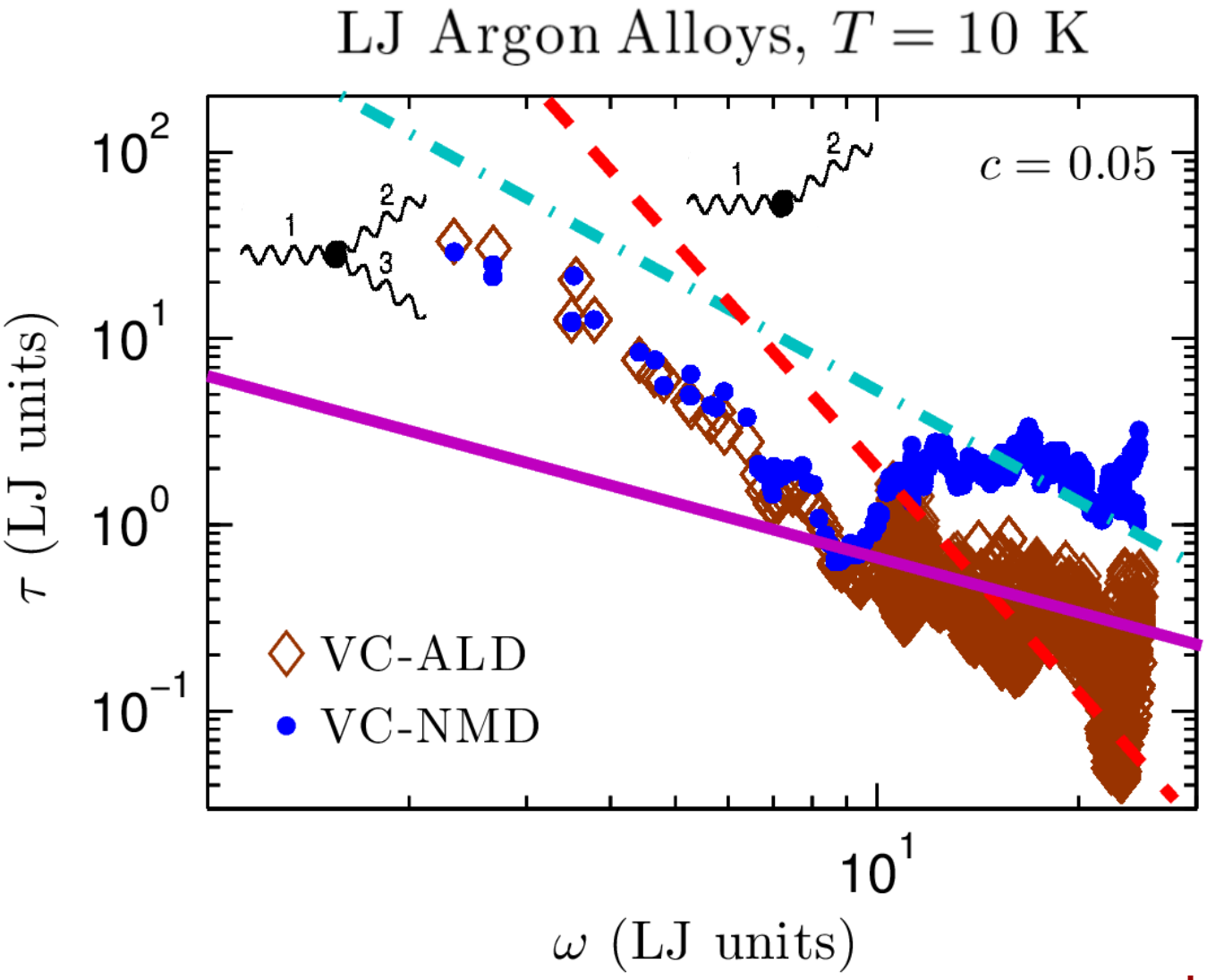
Gamma





VC-NMD vs VC-ALD

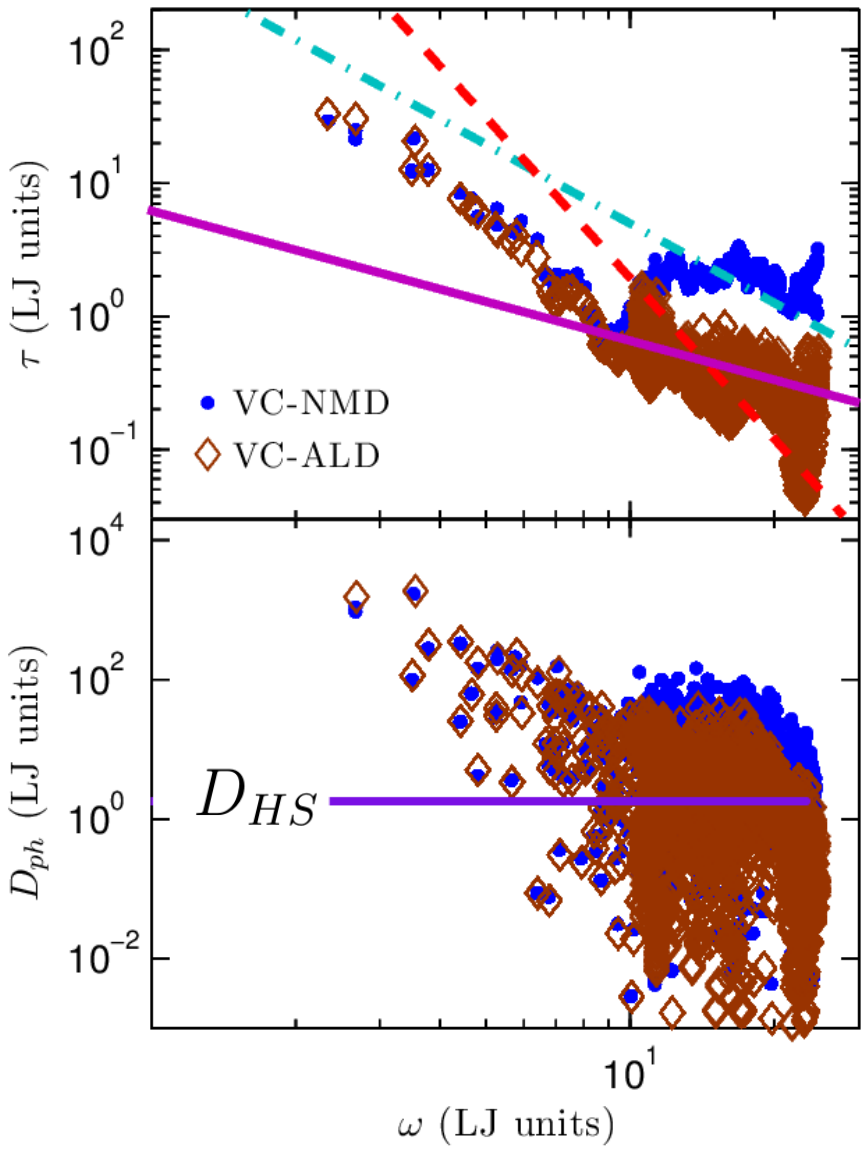




VC Diffusivities

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

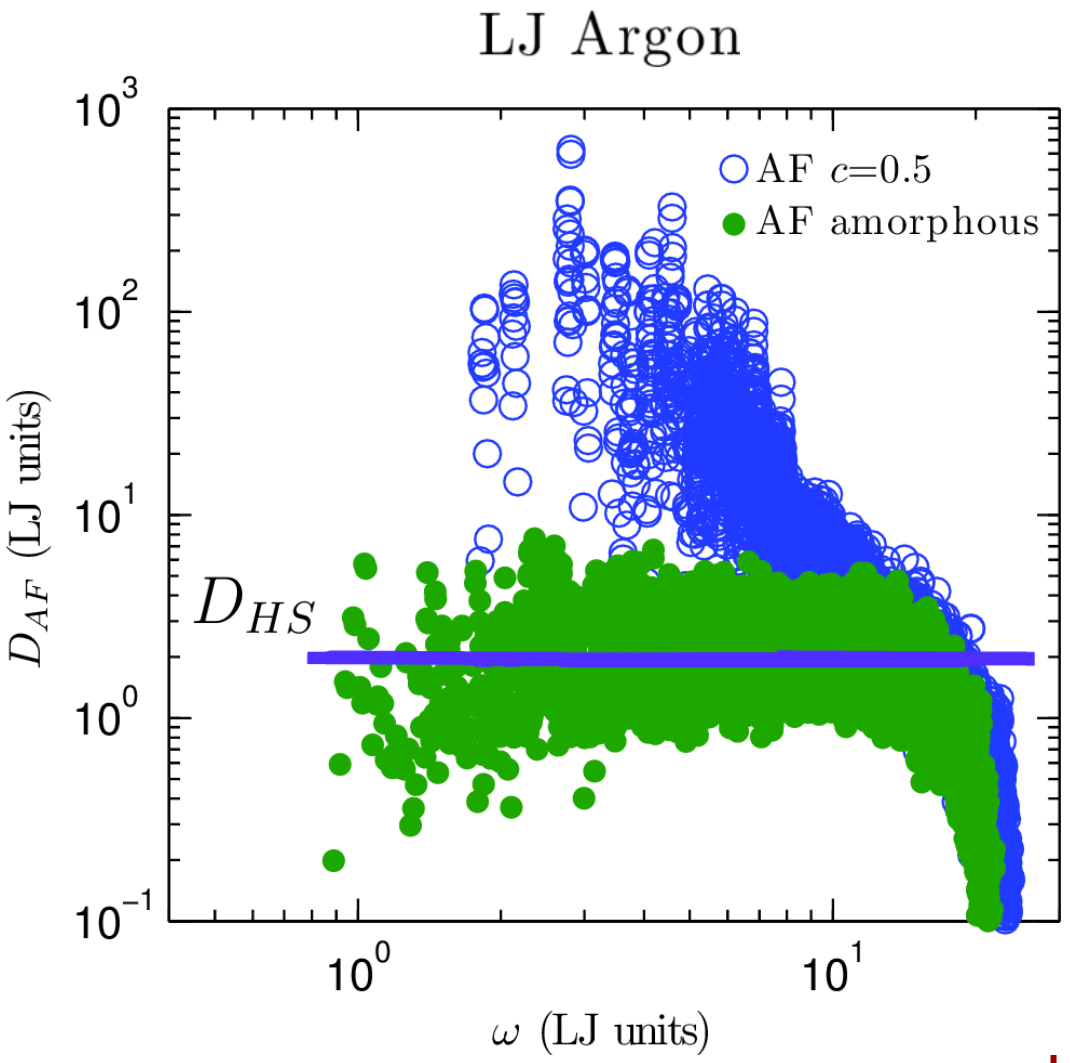
$$D_{HS} = \frac{1}{3} v_s a$$



AF Diffusivities

Allen-Feldman (**AF**) Theory:

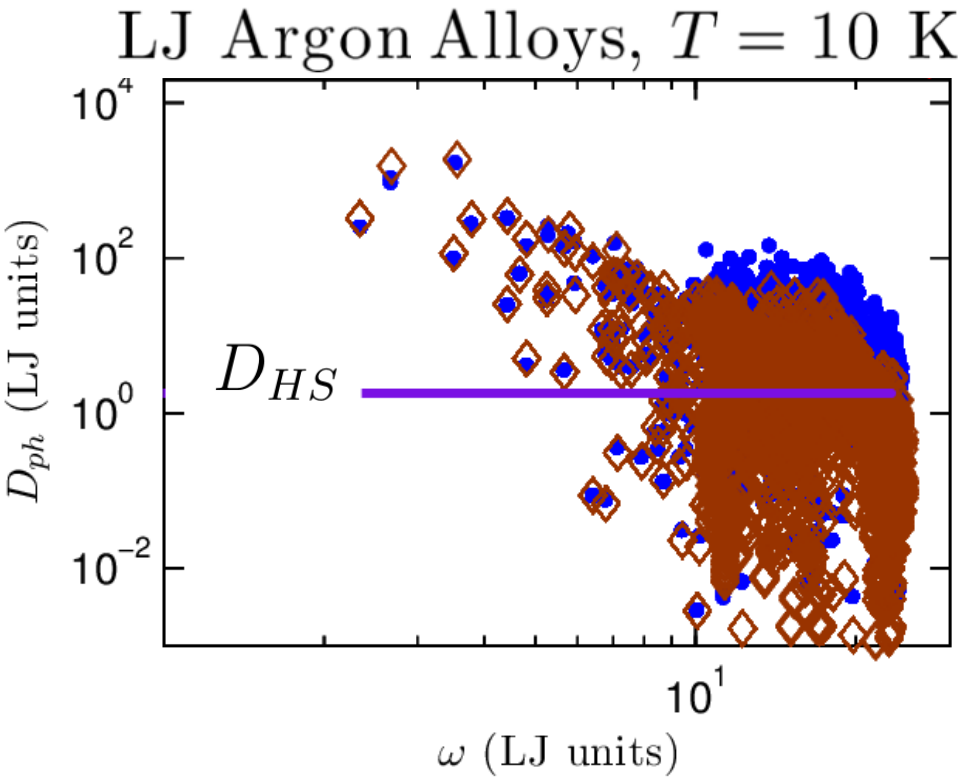
$$k_{AF} = \sum_{diffusons} \frac{k_B}{V} D_{AF,i}(\omega_i)$$



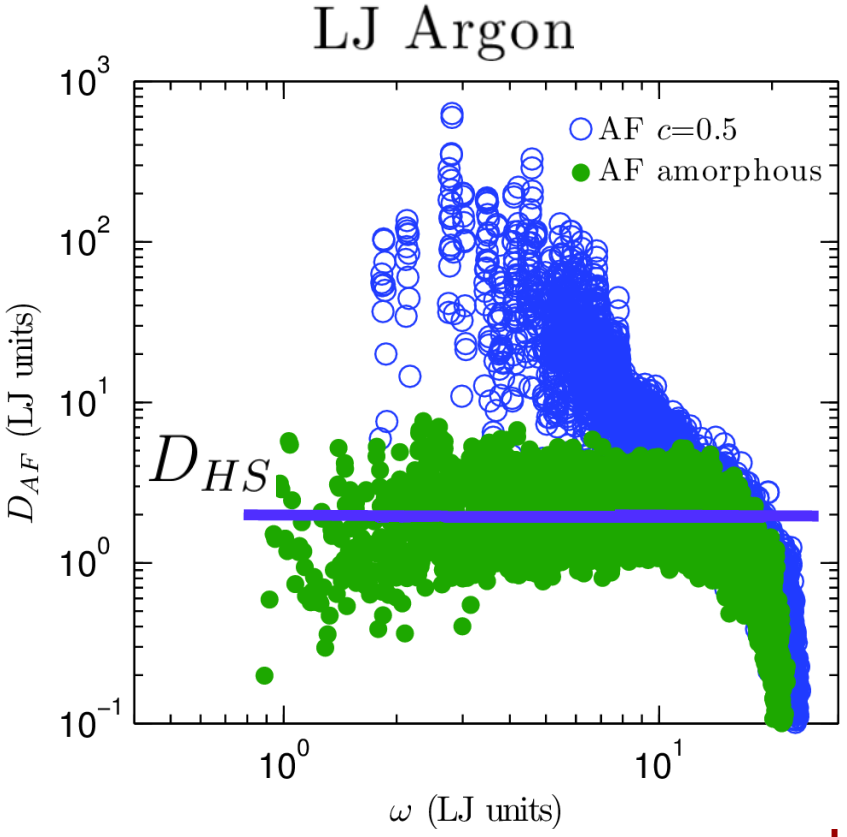
Physical Review B 48, 1258112588 (1993)

AF and VC Diffusivities

Phonons



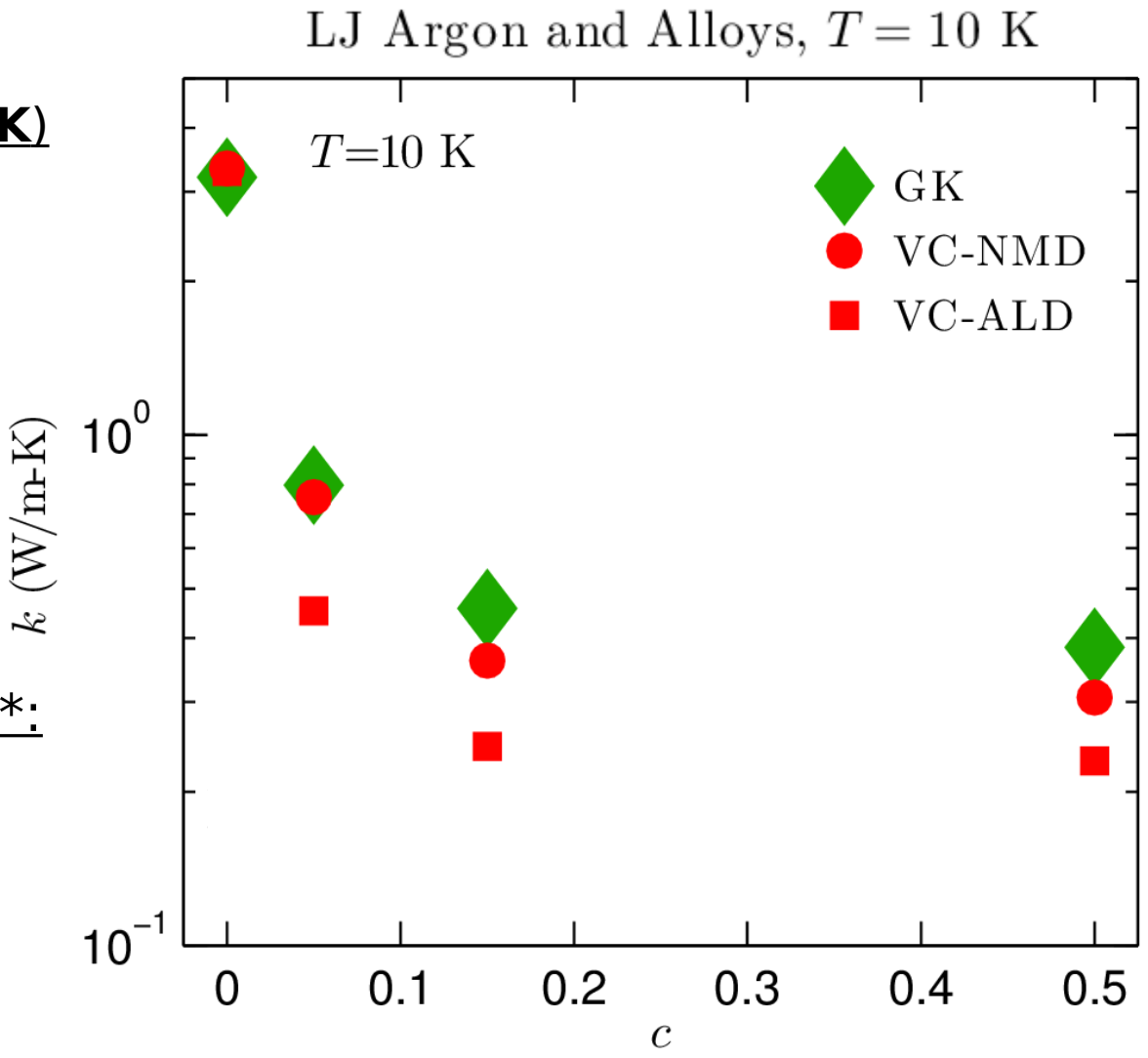
Diffusons



Physical Review B 48, 1258112588 (1993)

Thermal conductivity

MD-based Green-Kubo (**GK**)



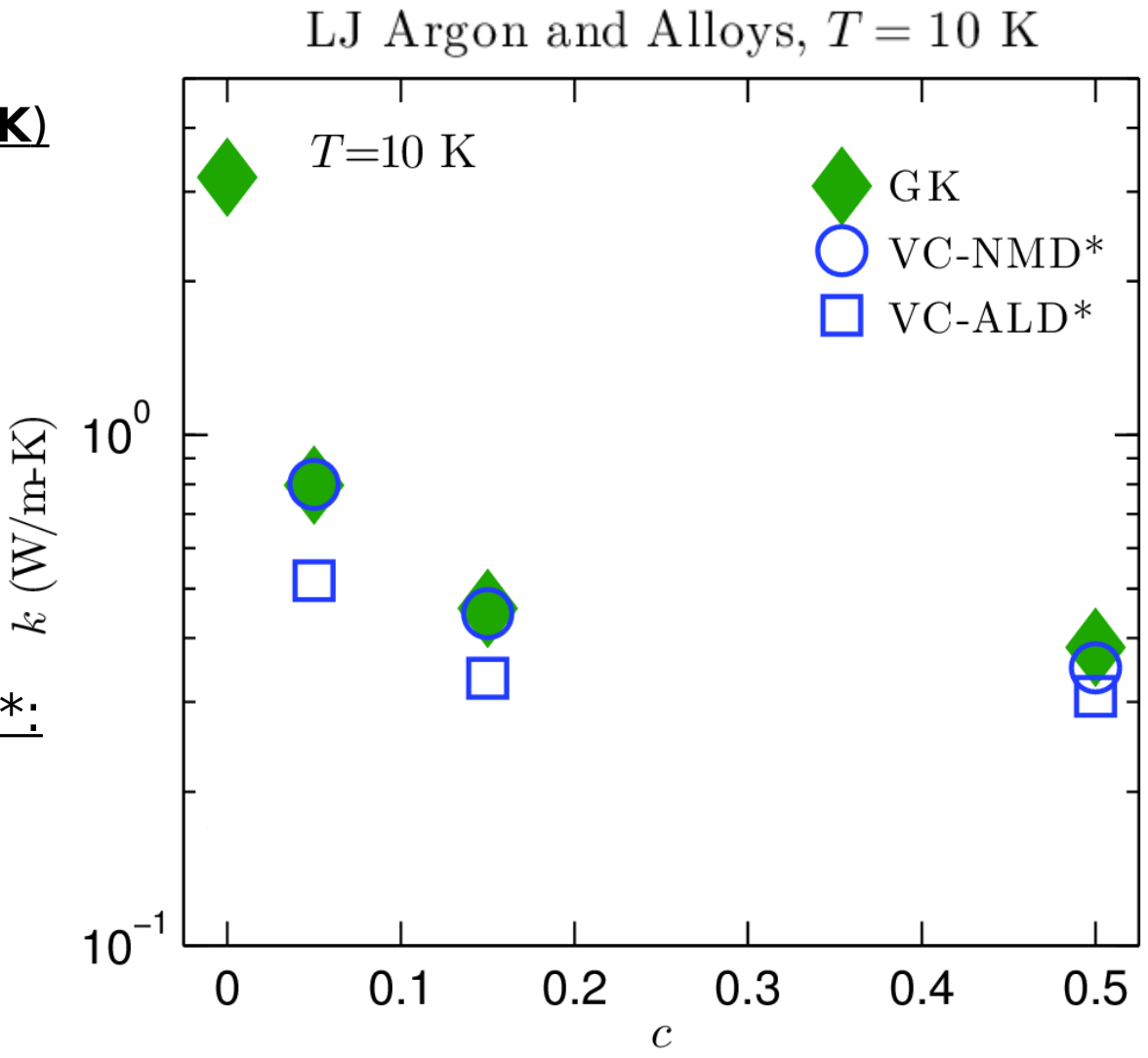
High-scatter adjustment*:

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

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

Thermal conductivity

MD-based Green-Kubo (**GK**)

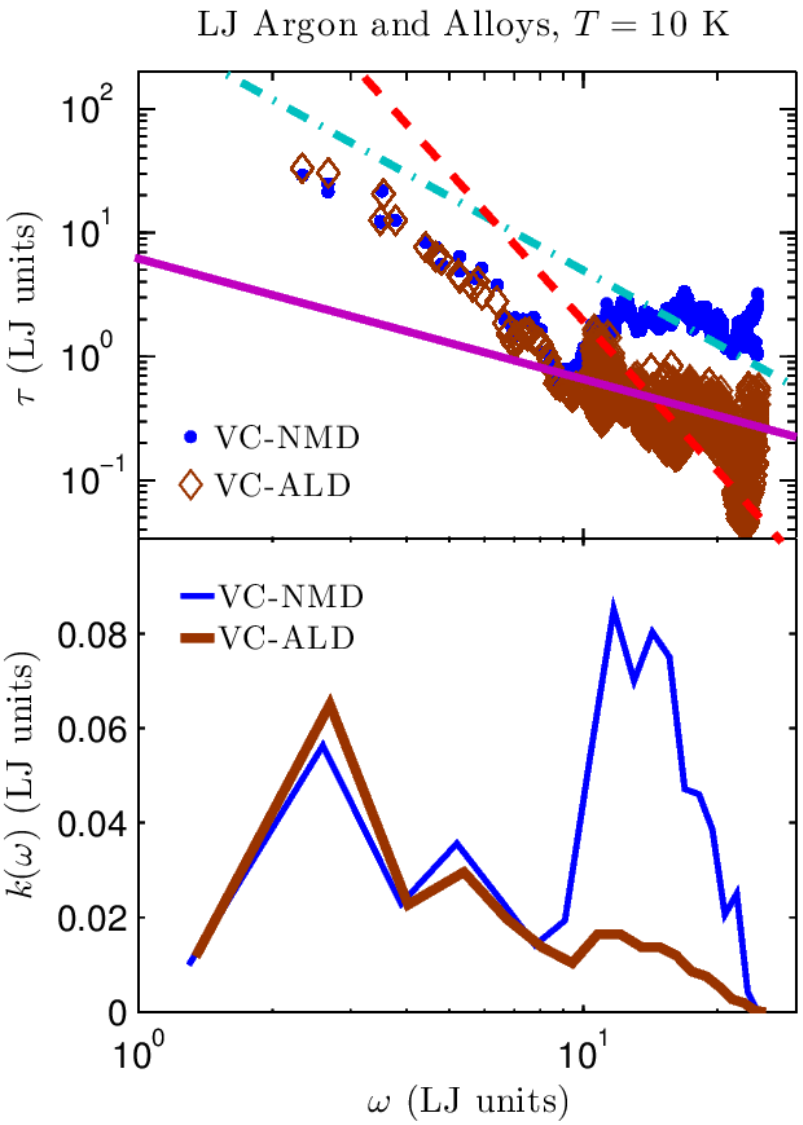


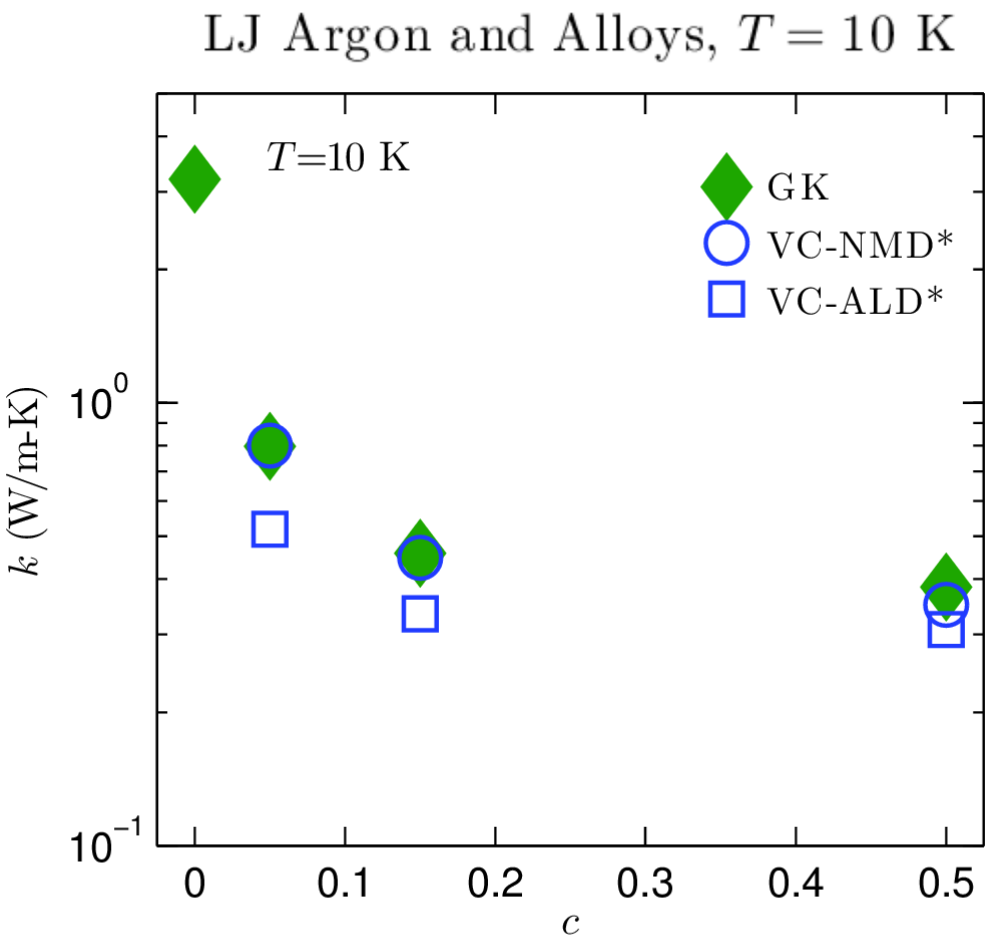
High-scatter adjustment*:

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

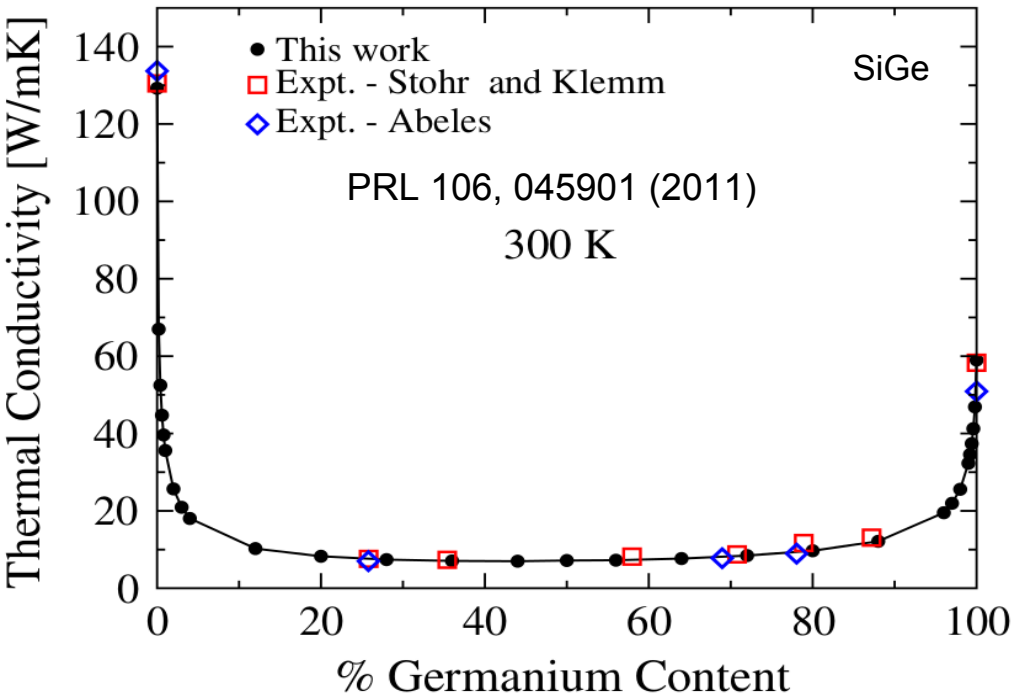
$$D_{ph}\binom{\kappa}{\nu} = D_{HS}$$

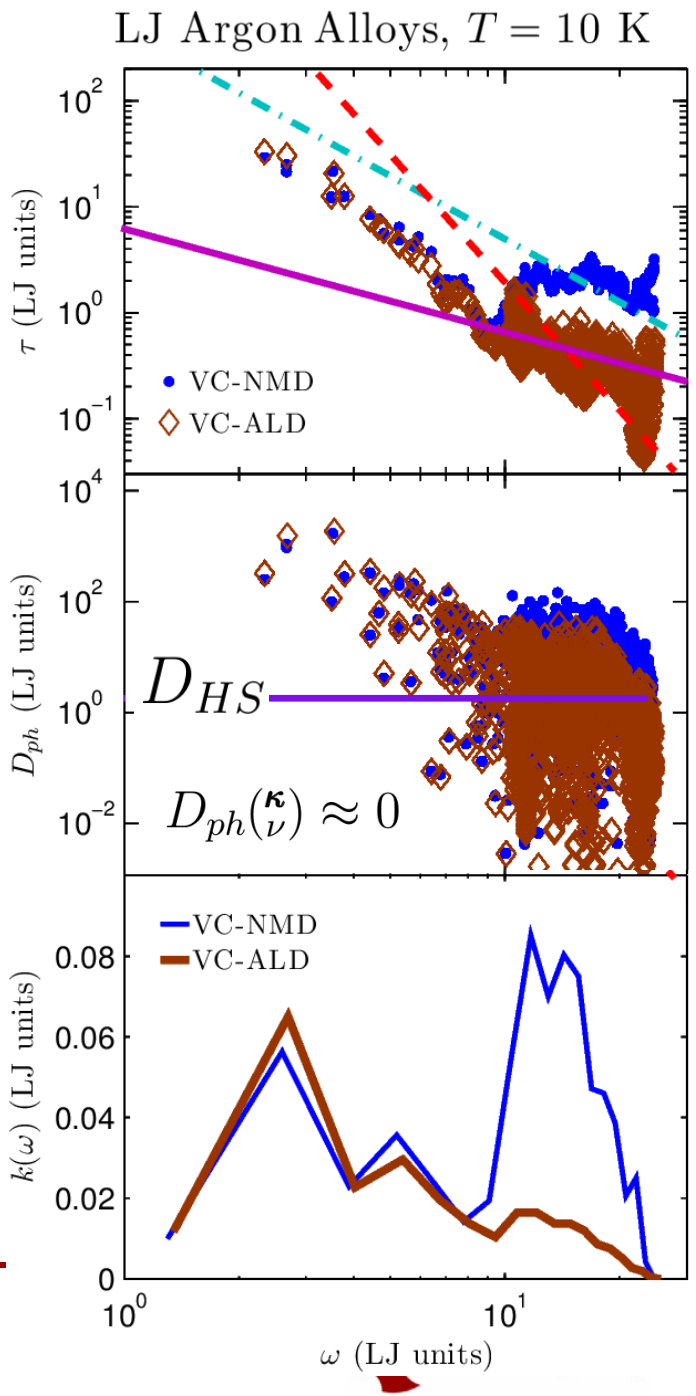
Thermal conductivity spectrum





Summary



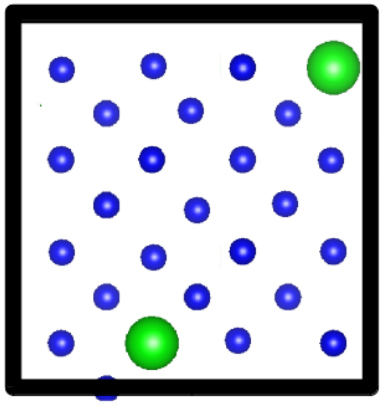


This work was supported by AFOSR award FA95501010098 and by a grant of computer time from the DOD High Performance Computing Modernization Program at the US Army Engineer Research and Development Center.

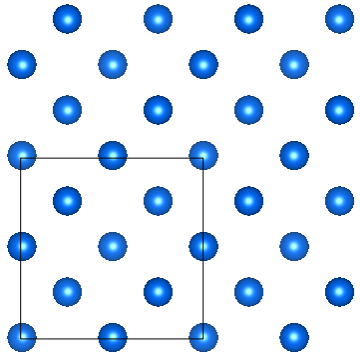
Carnegie Mellon

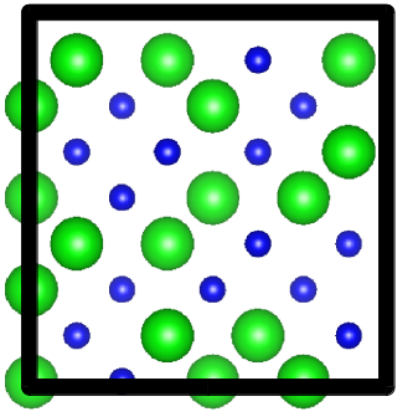
Explicit disorder: VC vs Gamma

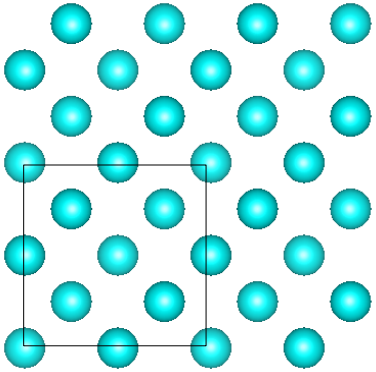
Gamma

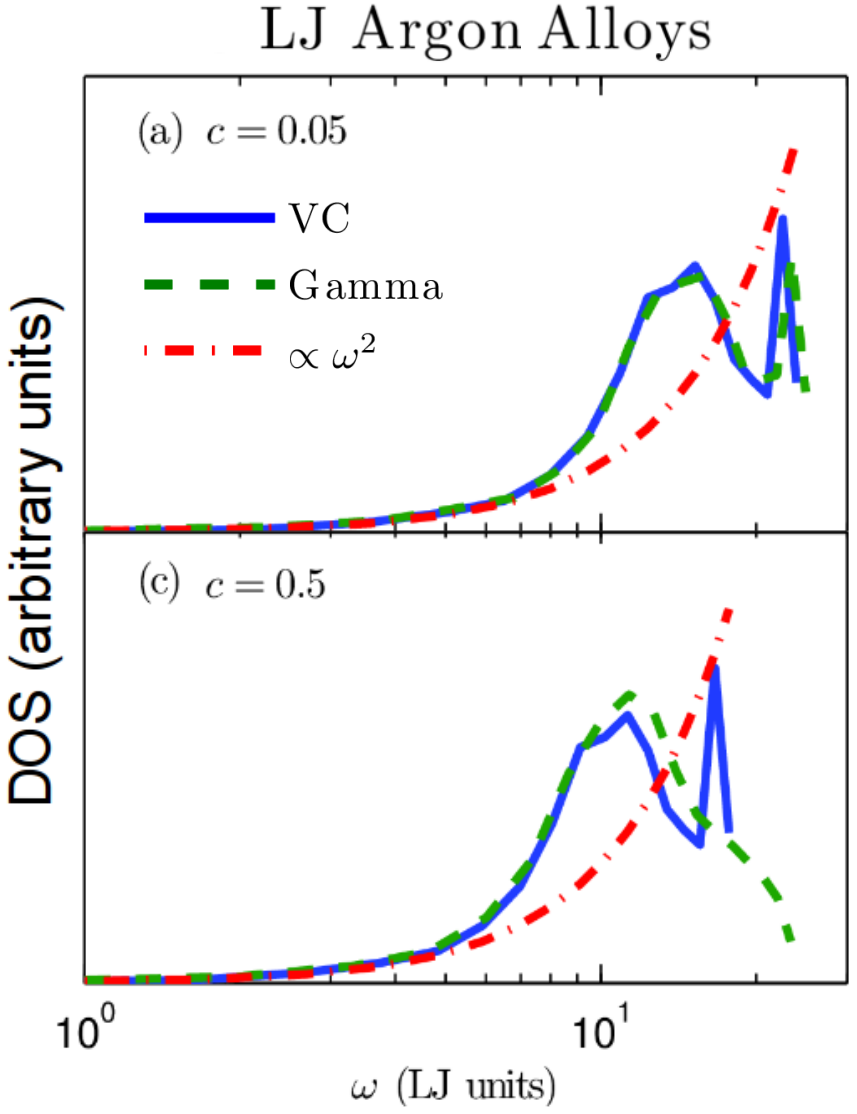


VC

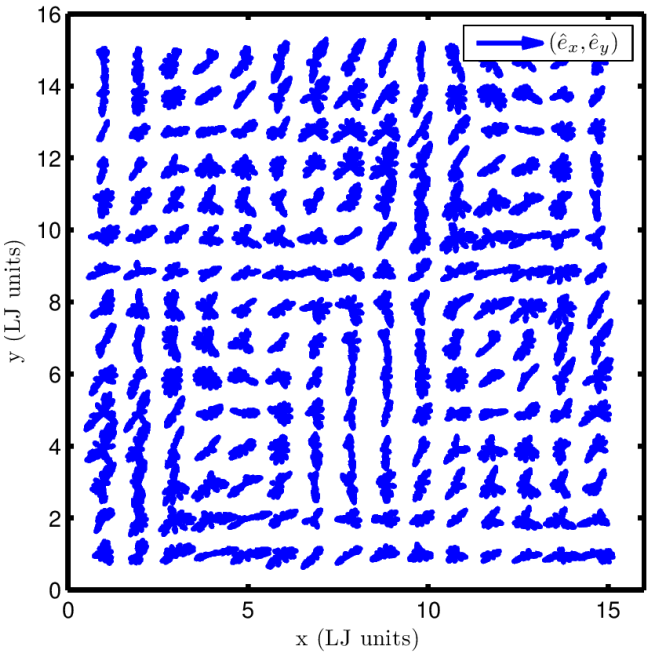


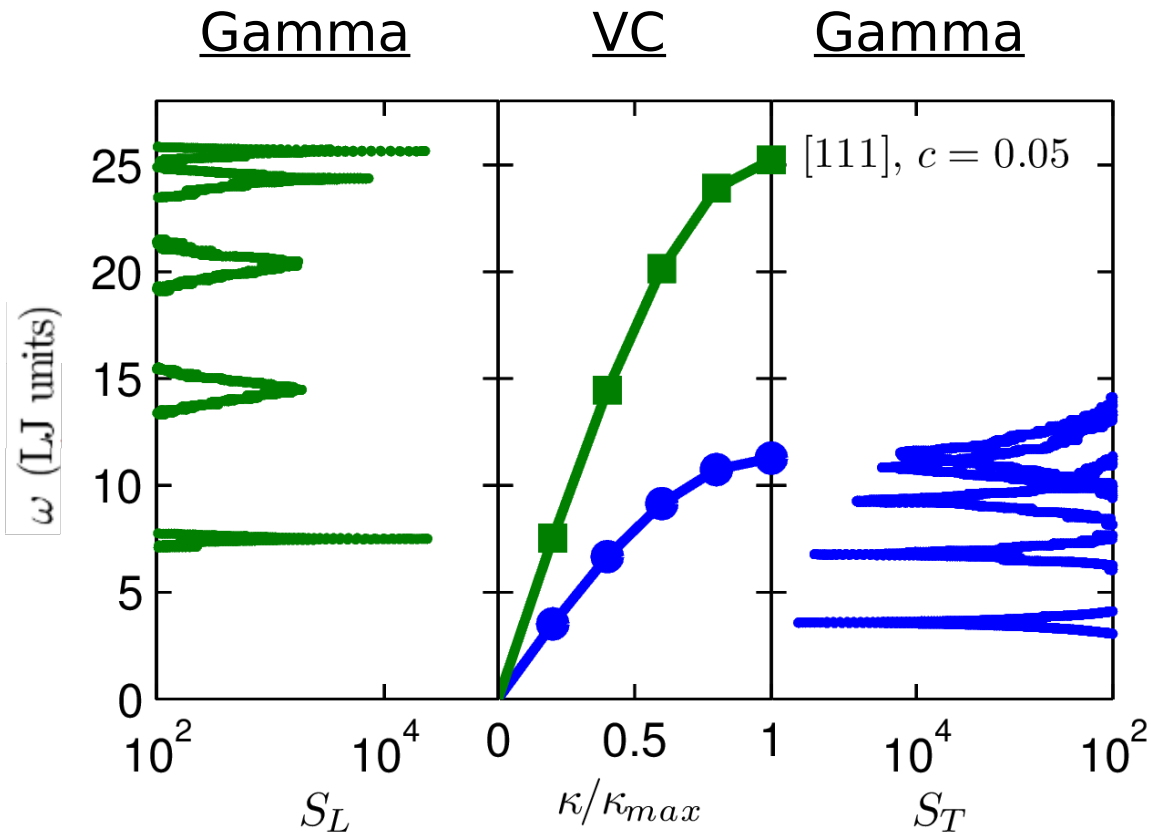






Explicit disorder: Structure Factor

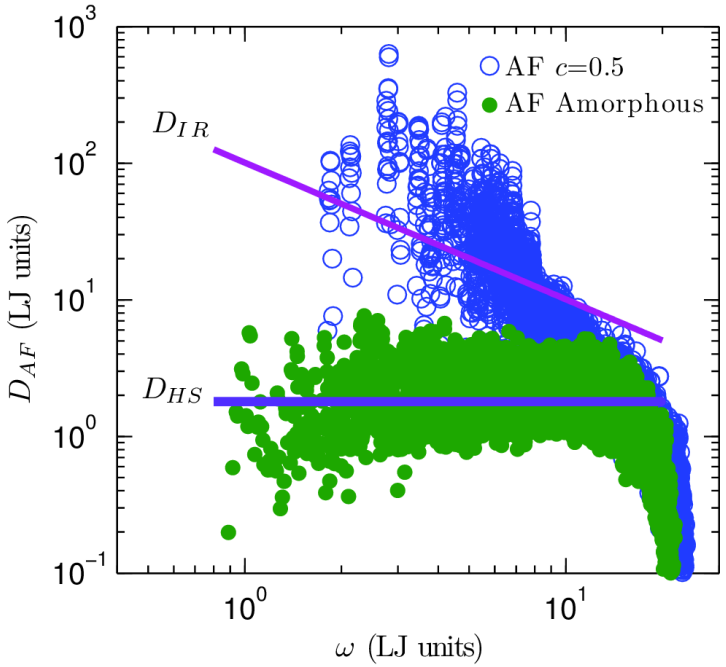


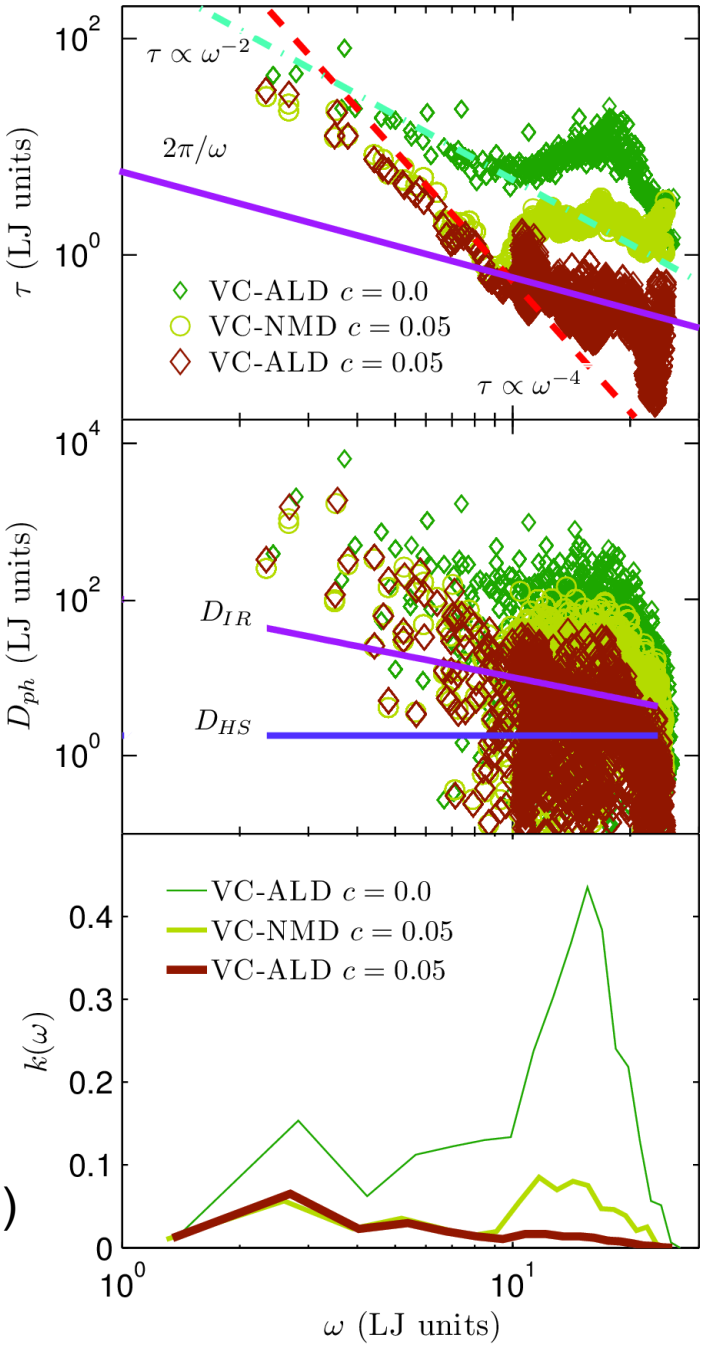


Phys. Rev. B 59, 35513559 (1999)

HS/IR Limit

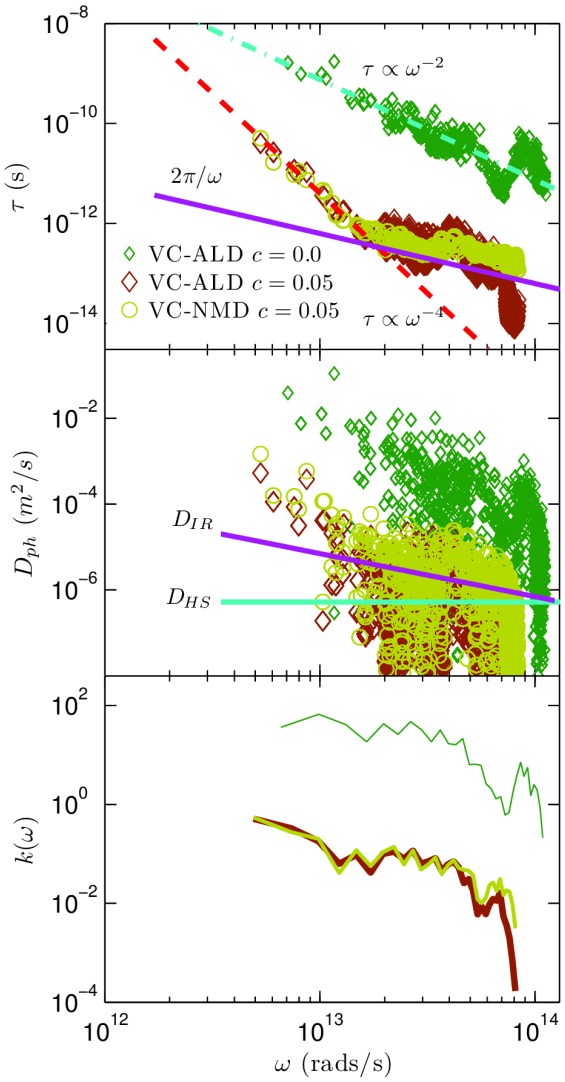
$$D_{IR} = \frac{2\pi}{3}\frac{v_s^2}{\omega}$$

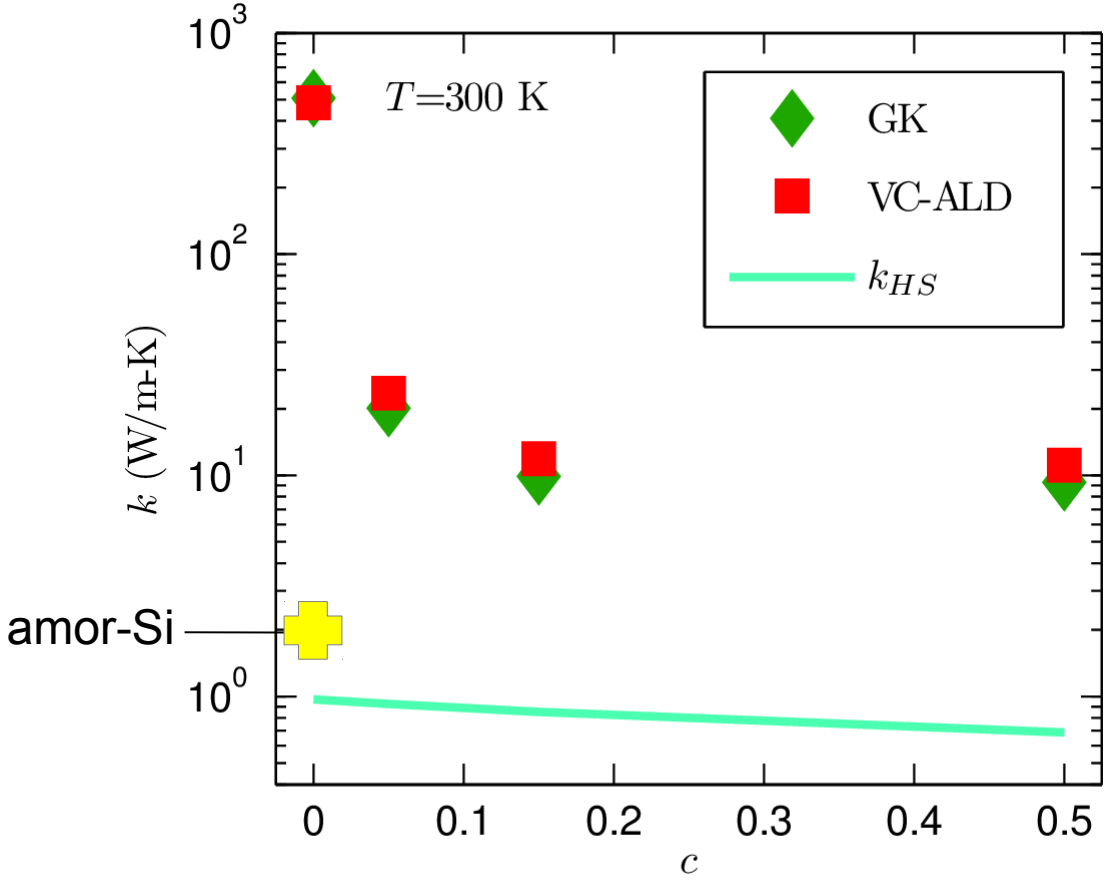




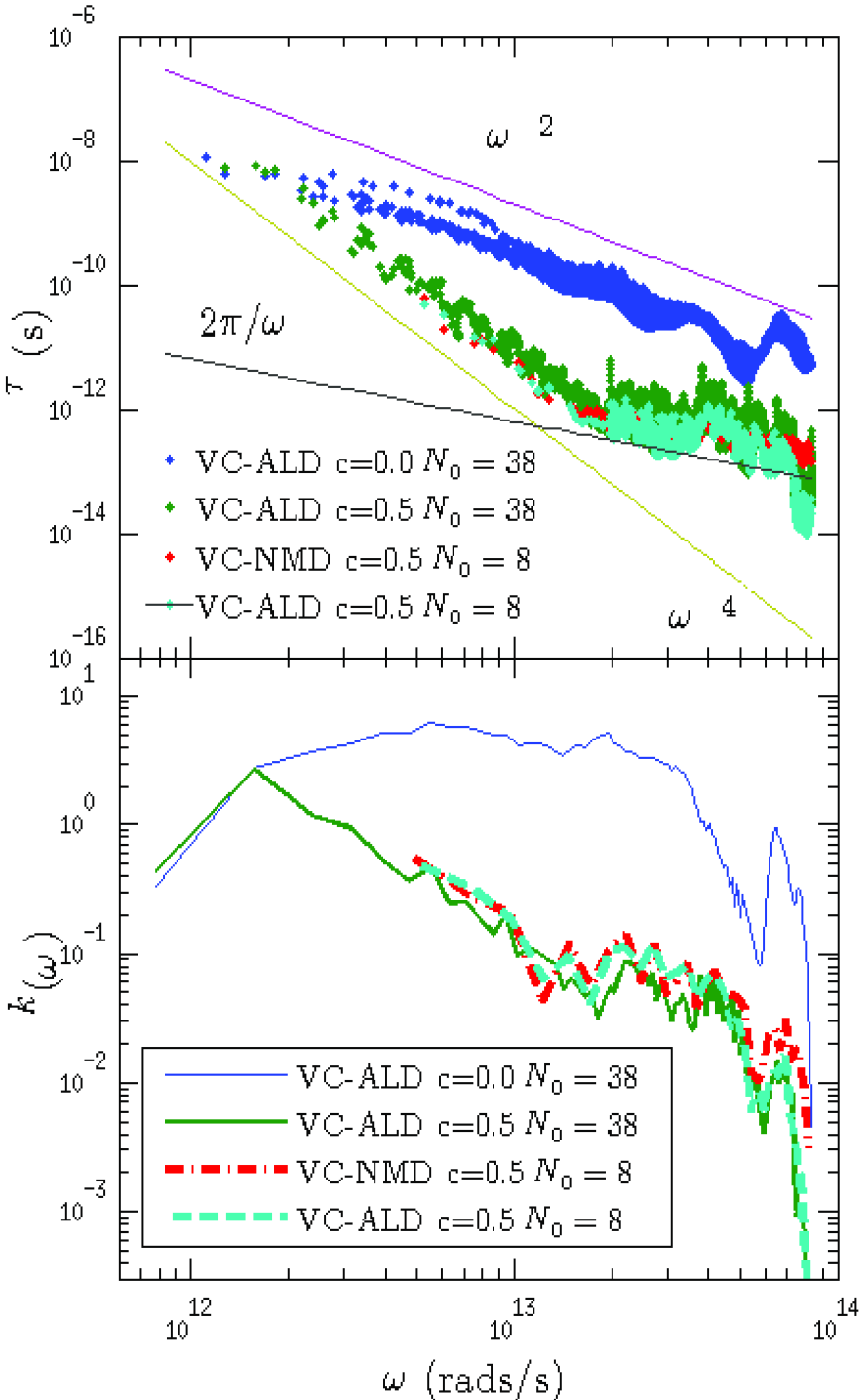
P. Sheng and M. Zhou, Science 253, 539542 (1991)

Thermal conductivity: SW silicon alloy





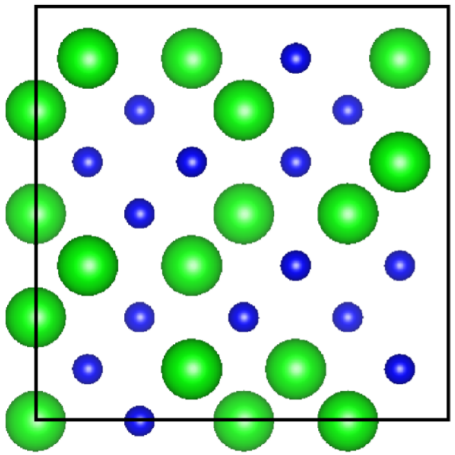
Phonon Spectrum: SW Si

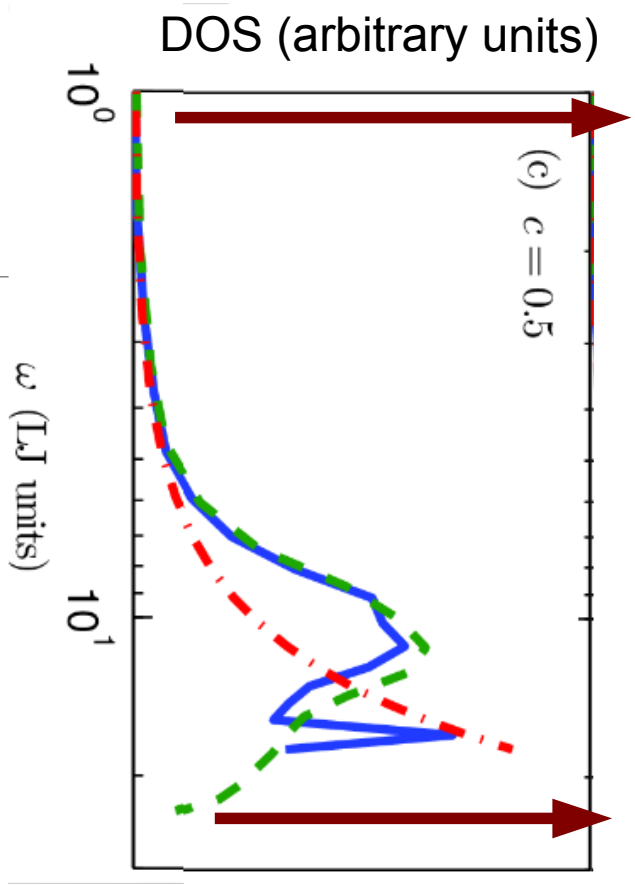


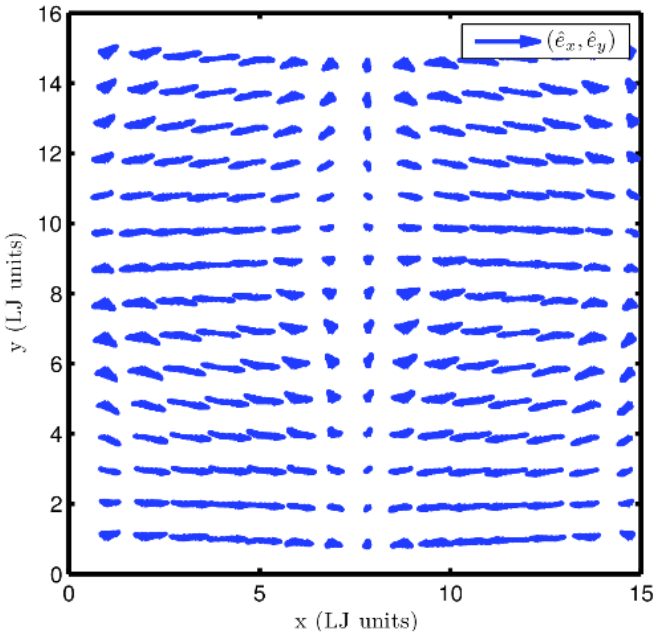
Gamma modes

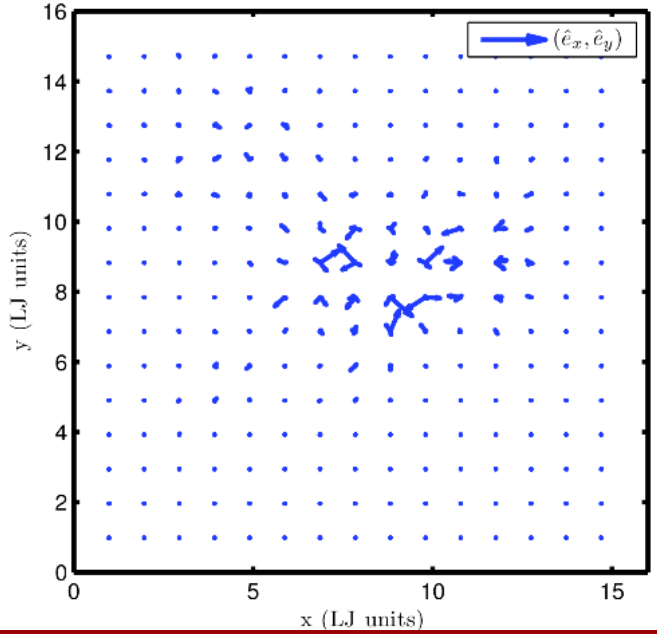
$e\left(\begin{smallmatrix}\kappa=0 & b\\ \nu & \alpha\end{smallmatrix}\right)$

(a) disordered supercell

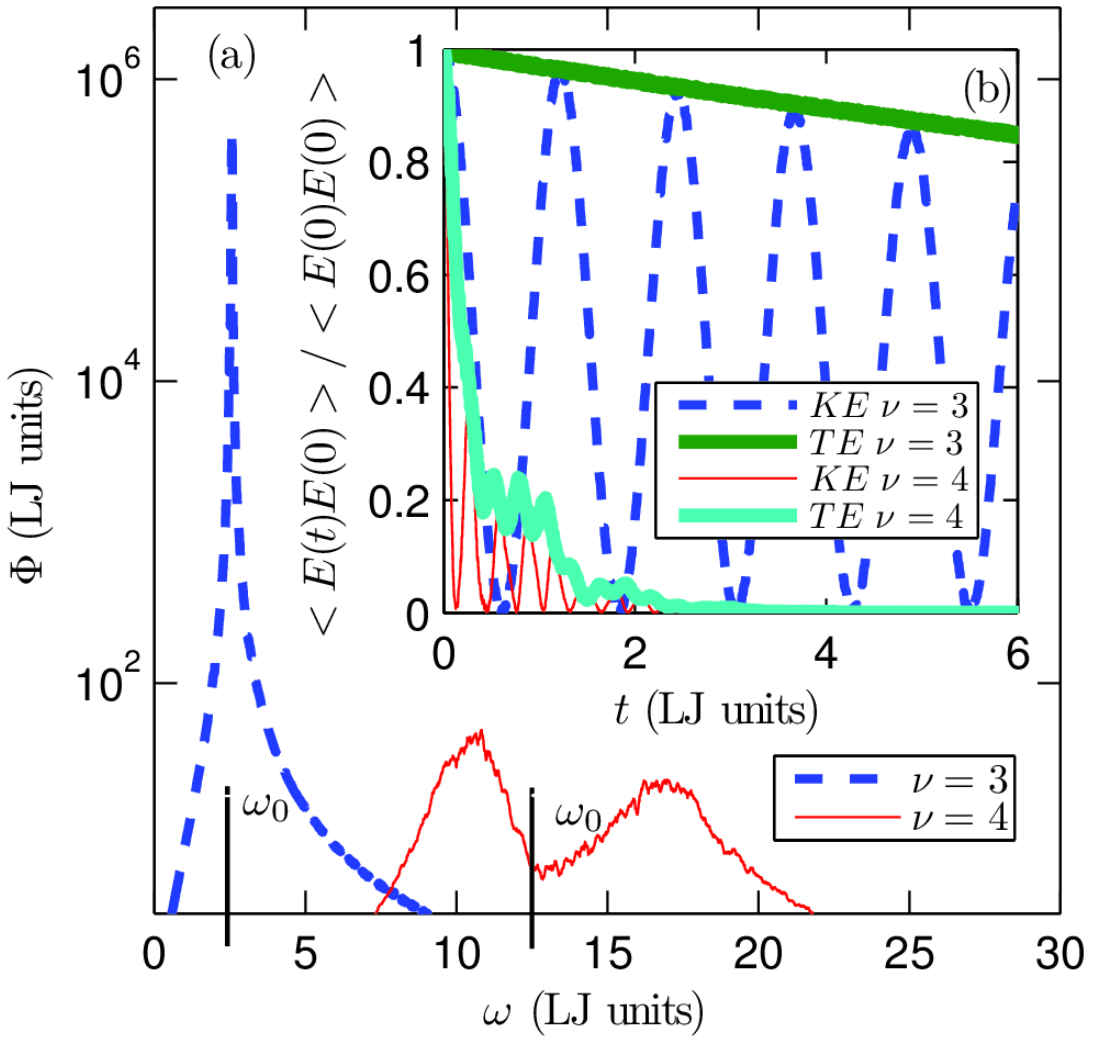




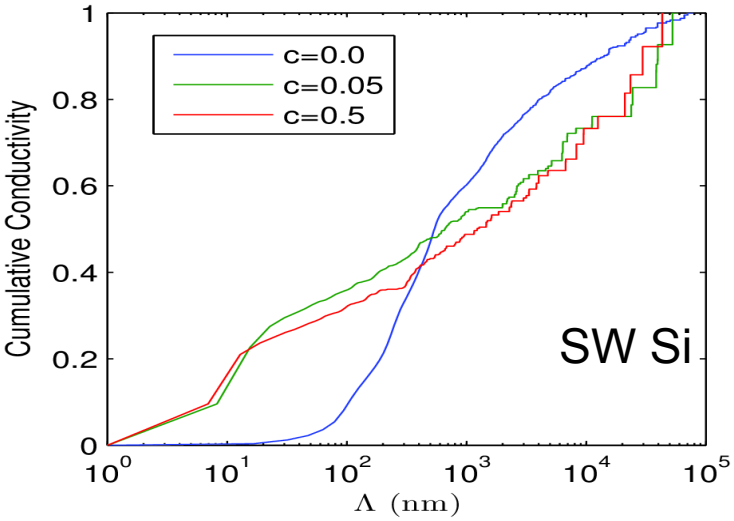


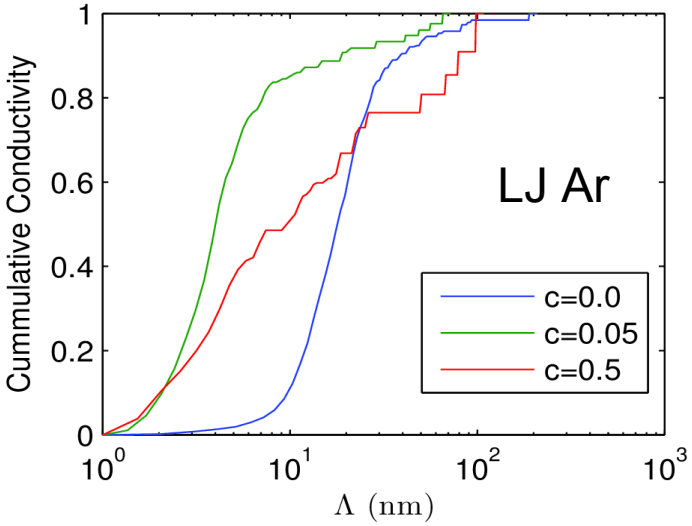


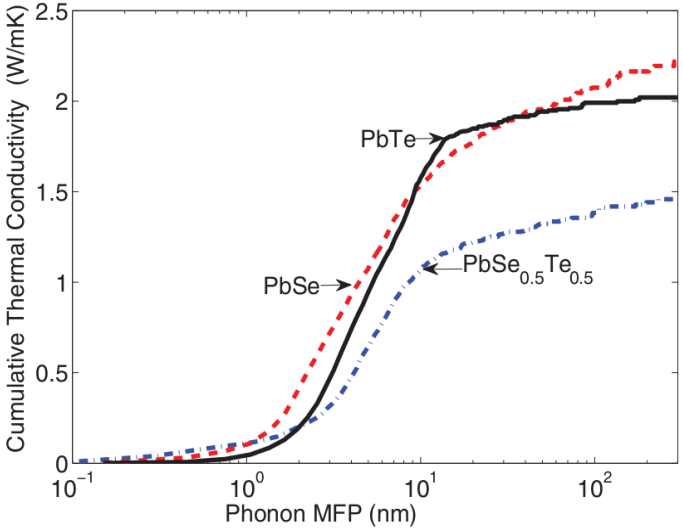
NMD using VC modes



Conductivity Accumulation







PHYSICAL REVIEW B 85, 184303 (2012)