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

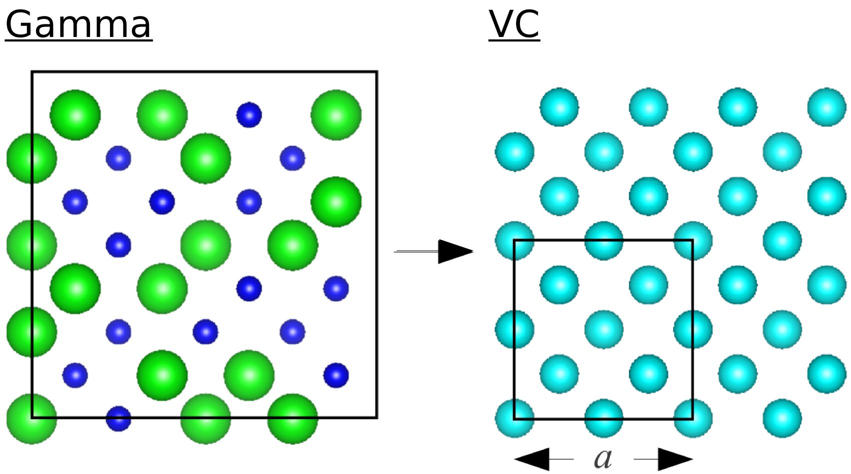
Jason Larkin and Alan J. H. McGaughey

Department of Mechanical Engineering
Carnegie Mellon University

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

04/04/13

Virtual Crystal Approximation



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

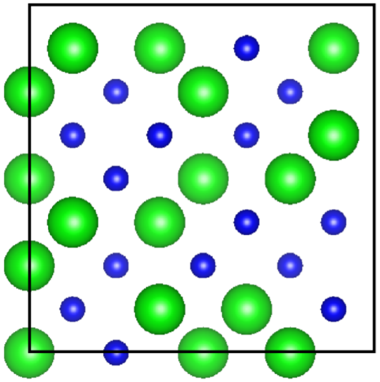
$$k_{ph,\mathbf{n}} = \sum_{\boldsymbol{\kappa}} \sum_{\nu} \frac{k_B}{V} D_{ph,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu}$$

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

Physical Review 131, 19061911 (1963)

Virtual Crystal Approximation

Gamma



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

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

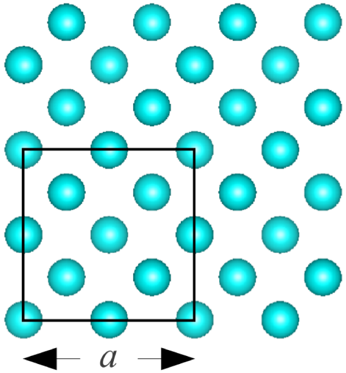
$$k_{AF} = \sum_{diffusons} \frac{k_B}{V} D_{AF,i}(\omega_i)$$

?

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

Physical Review B 48, 1258112588 (1993)

VC-ALD Diffusivities: Lifetimes



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

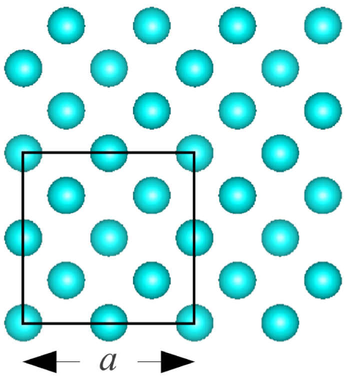
Perturbation theory:

Anharmonic Lattice Dynamics (**ALD**)

Matthiessen's Rule:

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

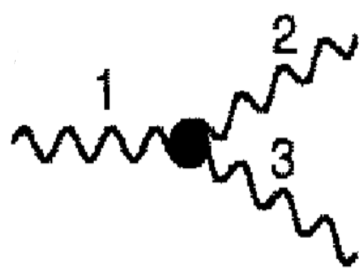
VC-ALD Diffusivities: Lifetimes



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

Perturbation theory:

Anharmonic Lattice Dynamics (**ALD**)



Phonon-Defect[1]

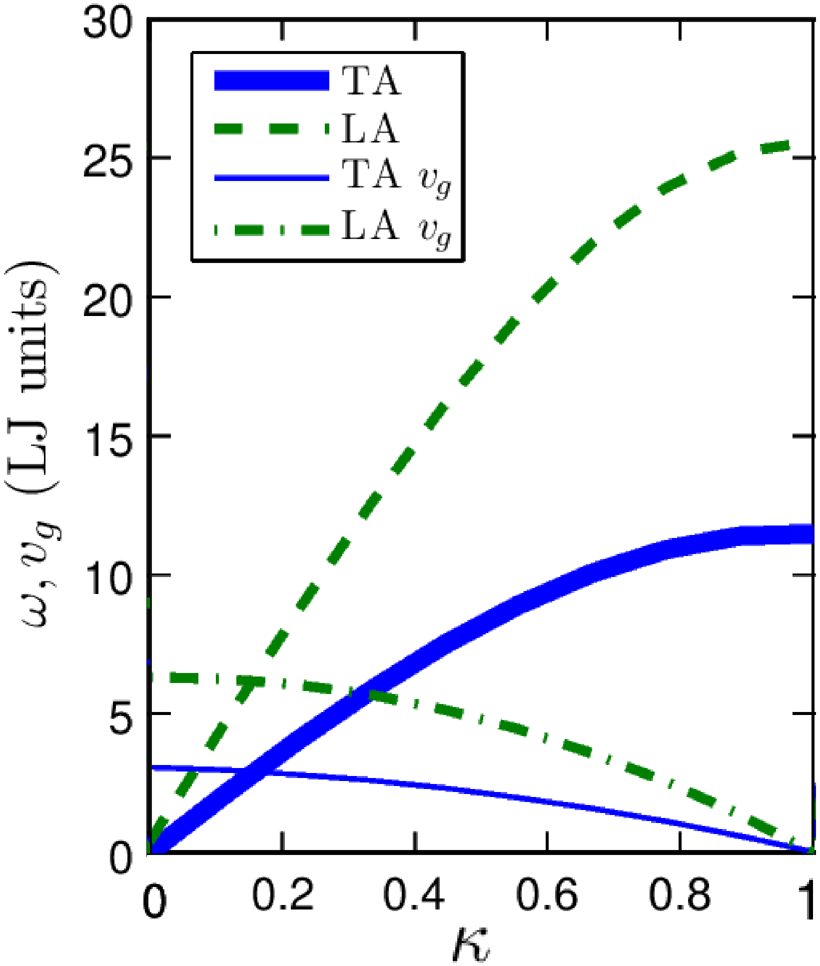
Matthiessen's Rule:

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

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

VC-ALD Diffusivities: Group Velocity

$$\mathbf{v}_{g,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = \frac{\partial \omega\binom{\boldsymbol{\kappa}}{\nu}}{\partial \boldsymbol{\kappa}}$$



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

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

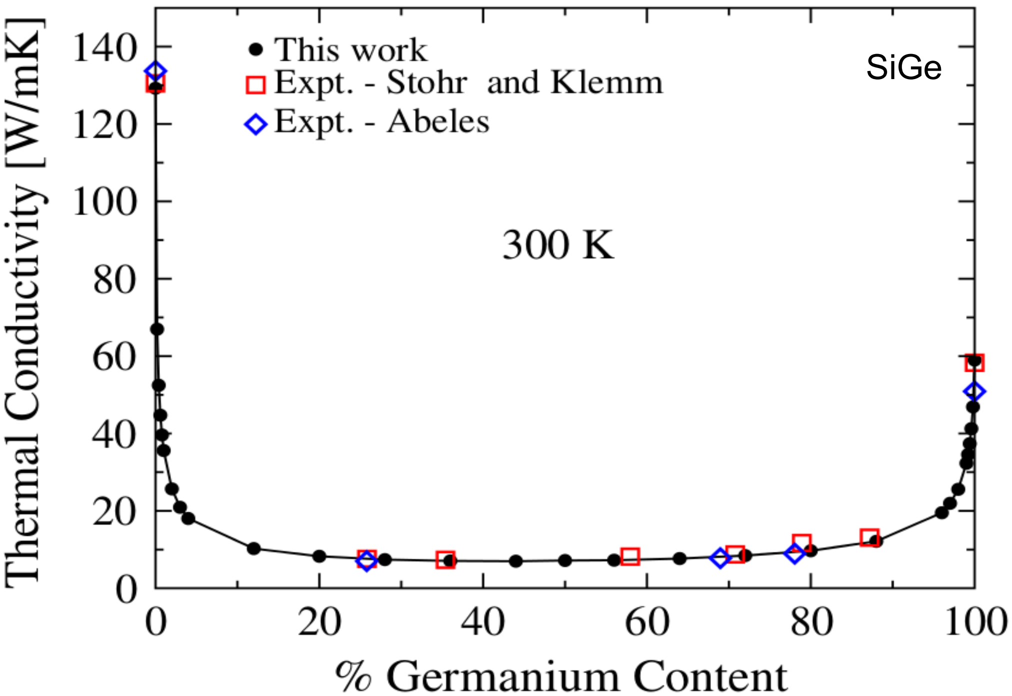
High-Scatter limit:

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

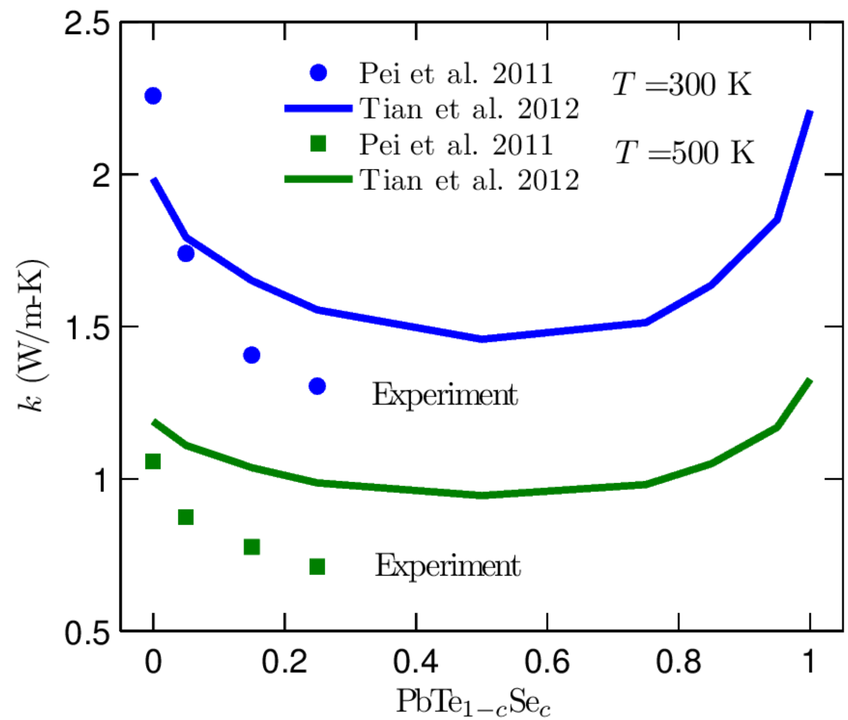
[1] Ann. Rev. of Phys. Chem. 39, 93121 (1988)

VC-ALD: experimental accuracy

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



PRL 106, 045901 (2011)

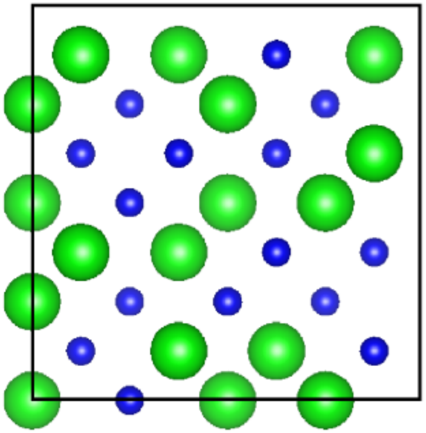


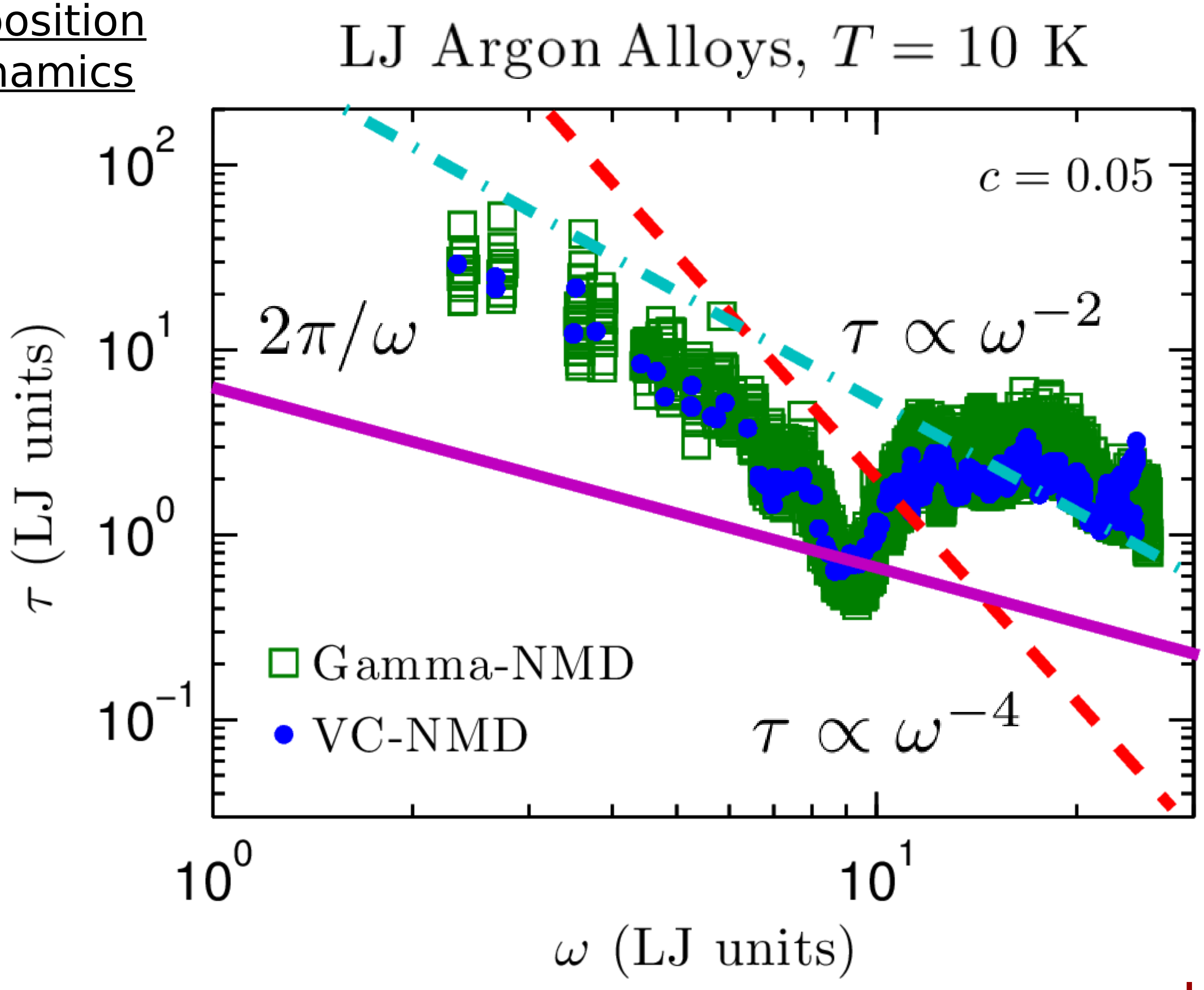
PRB 85, 184303 (2012)

Explicit disorder: NMD

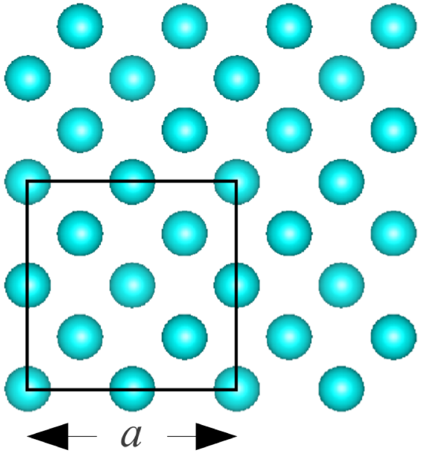
Normal Mode Decomposition (**NMD**): Molecular Dynamics

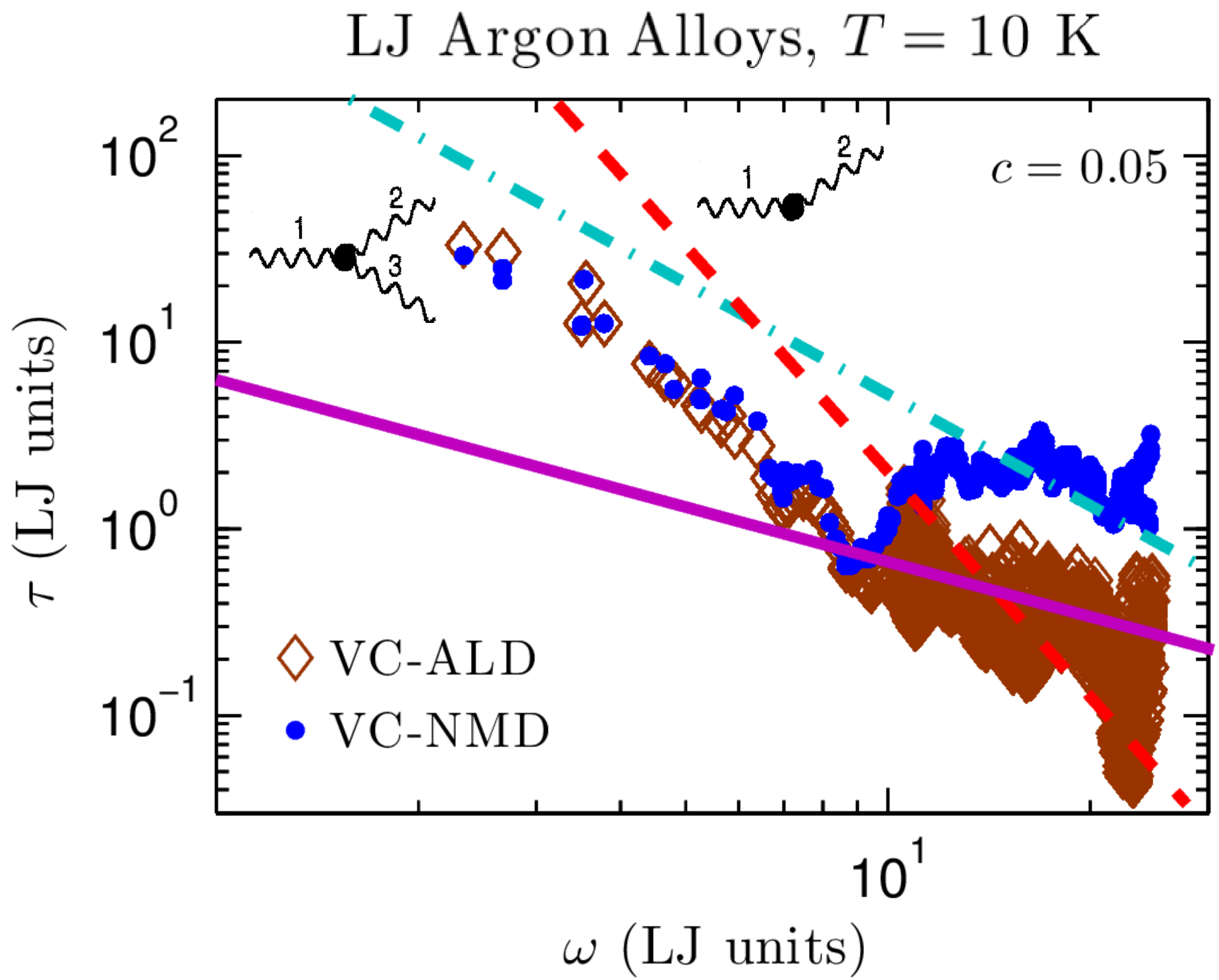
Disordered Supercell





VC-NMD vs VC-ALD

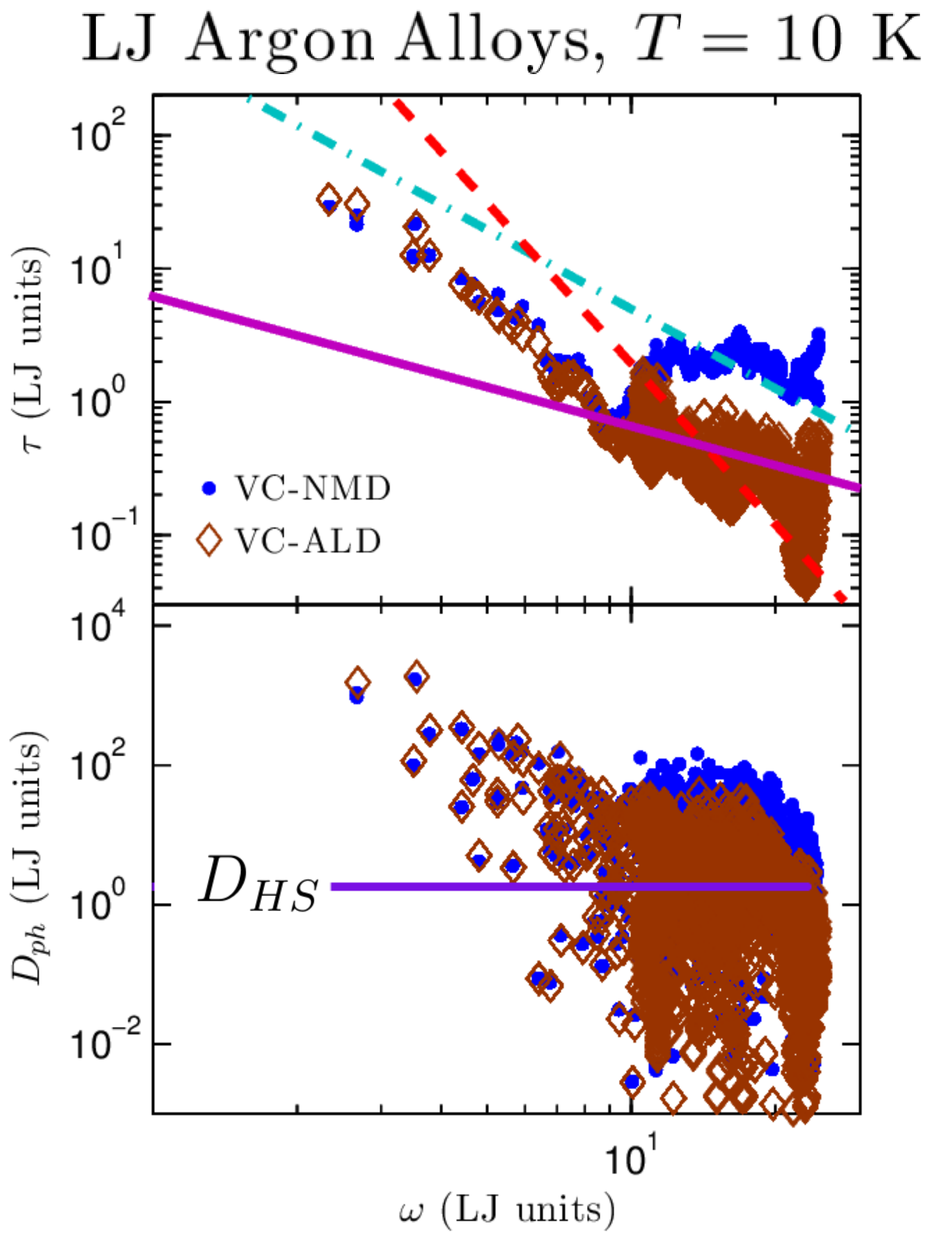




VC Diffusivities

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

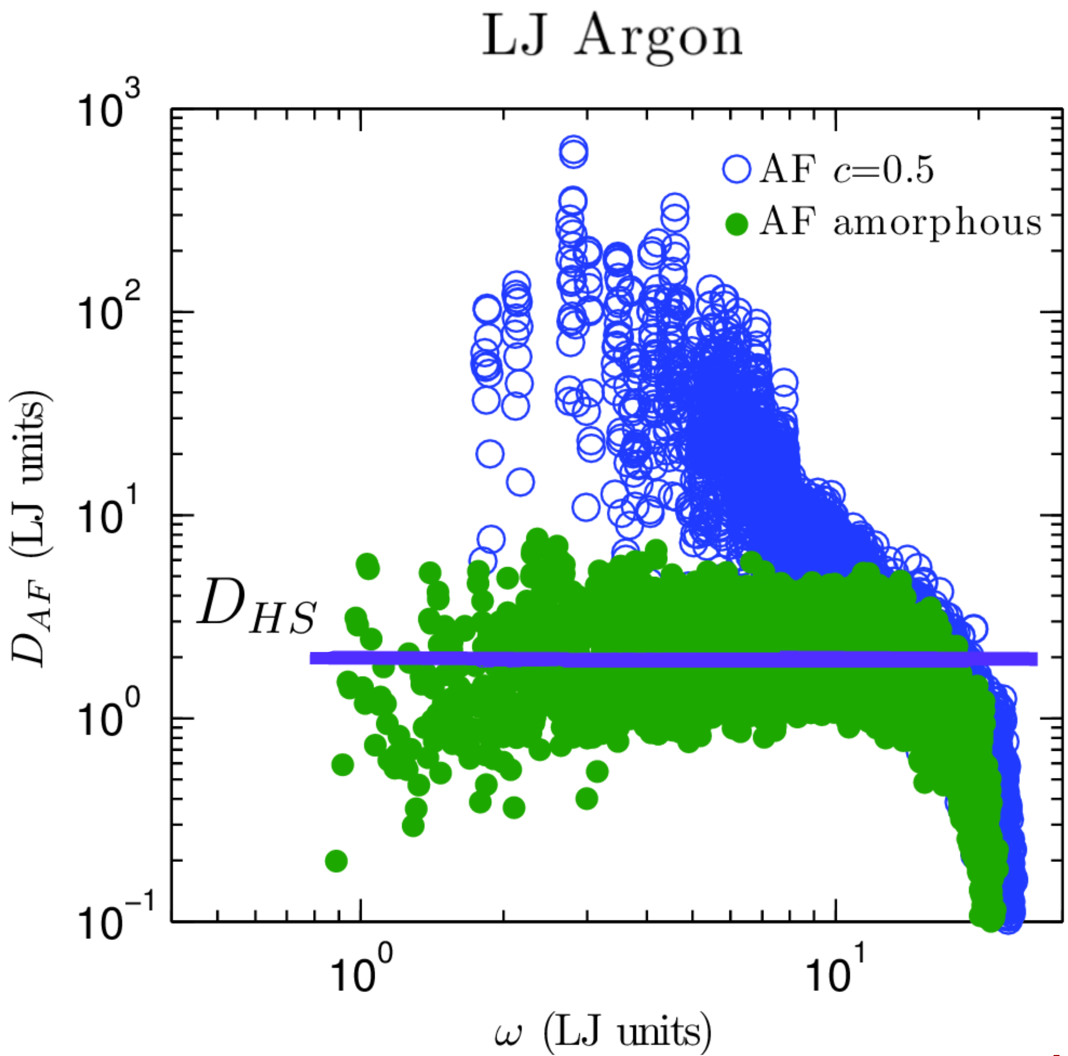
$$D_{HS} = \frac{1}{3} v_s a$$



AF Diffusivities

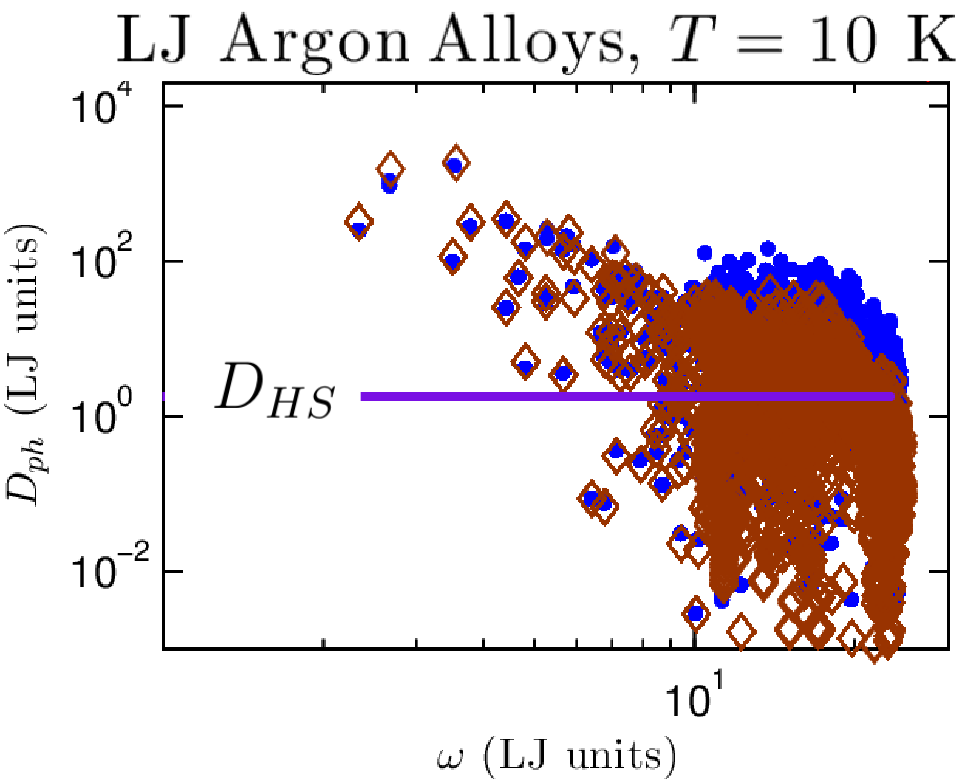
Allen-Feldman (**AF**) Theory:

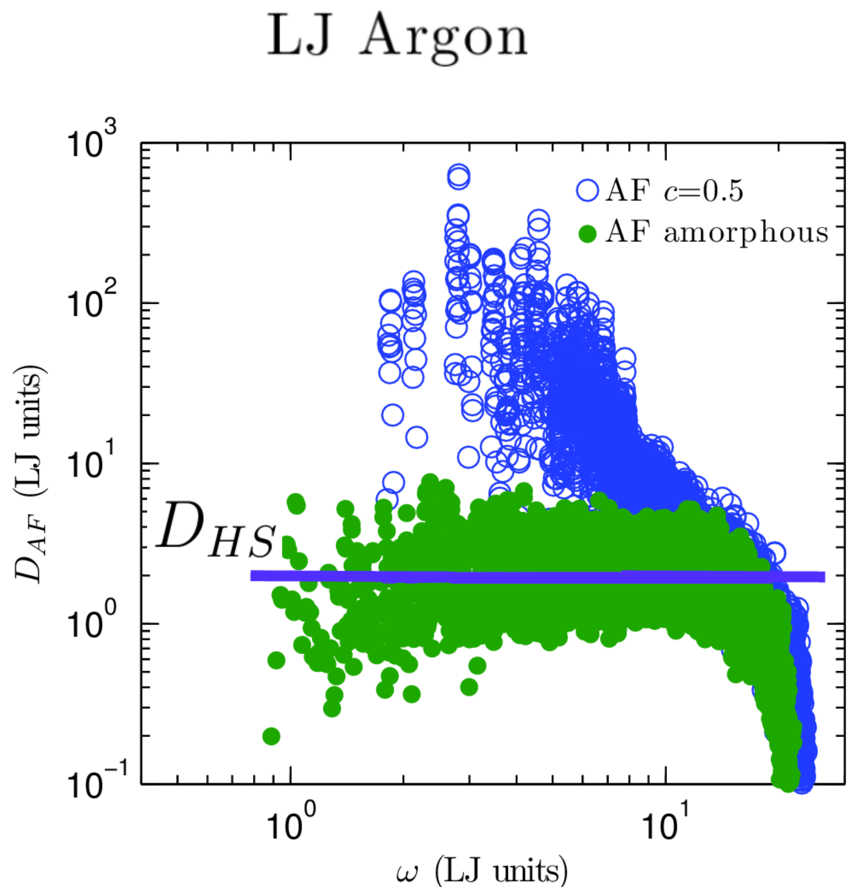
$$k_{AF} = \sum_{diffusons} \frac{k_B}{V} D_{AF,i}(\omega_i)$$



Physical Review B 48, 1258112588 (1993)

AF Diffusivities





Physical Review B 48, 1258112588 (1993)

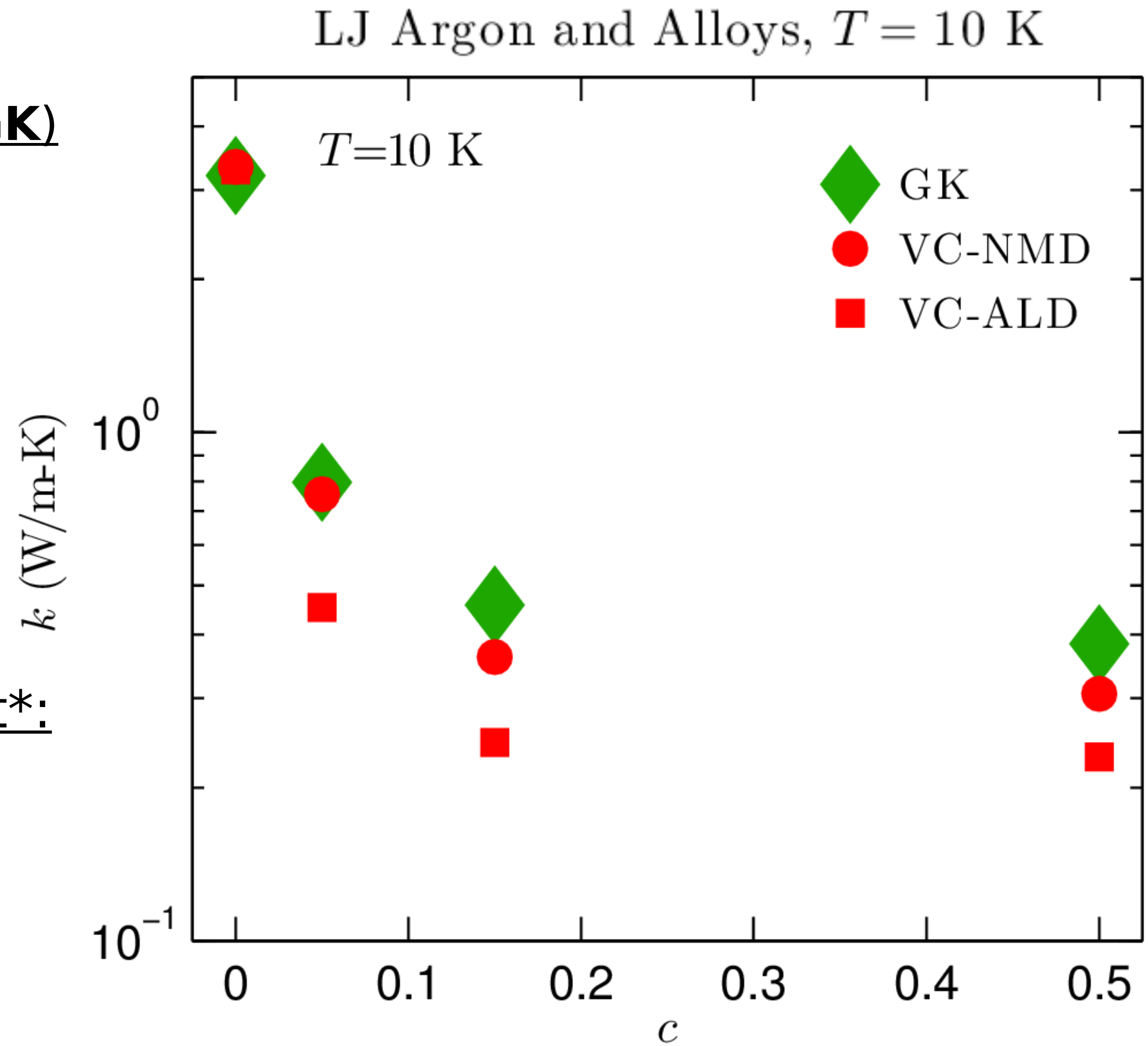
Thermal conductivity

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

High-scatter adjustment*:

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

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



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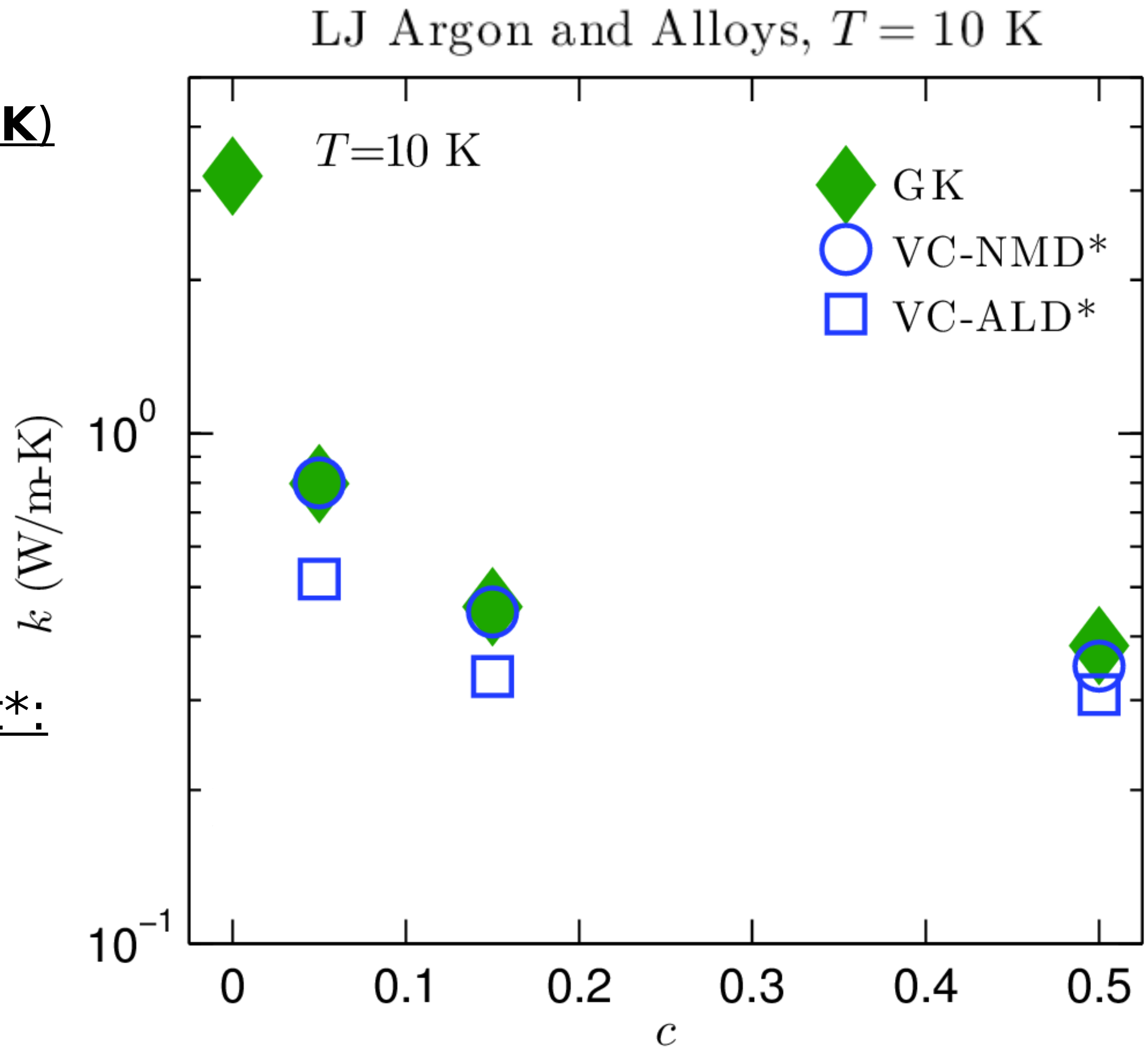
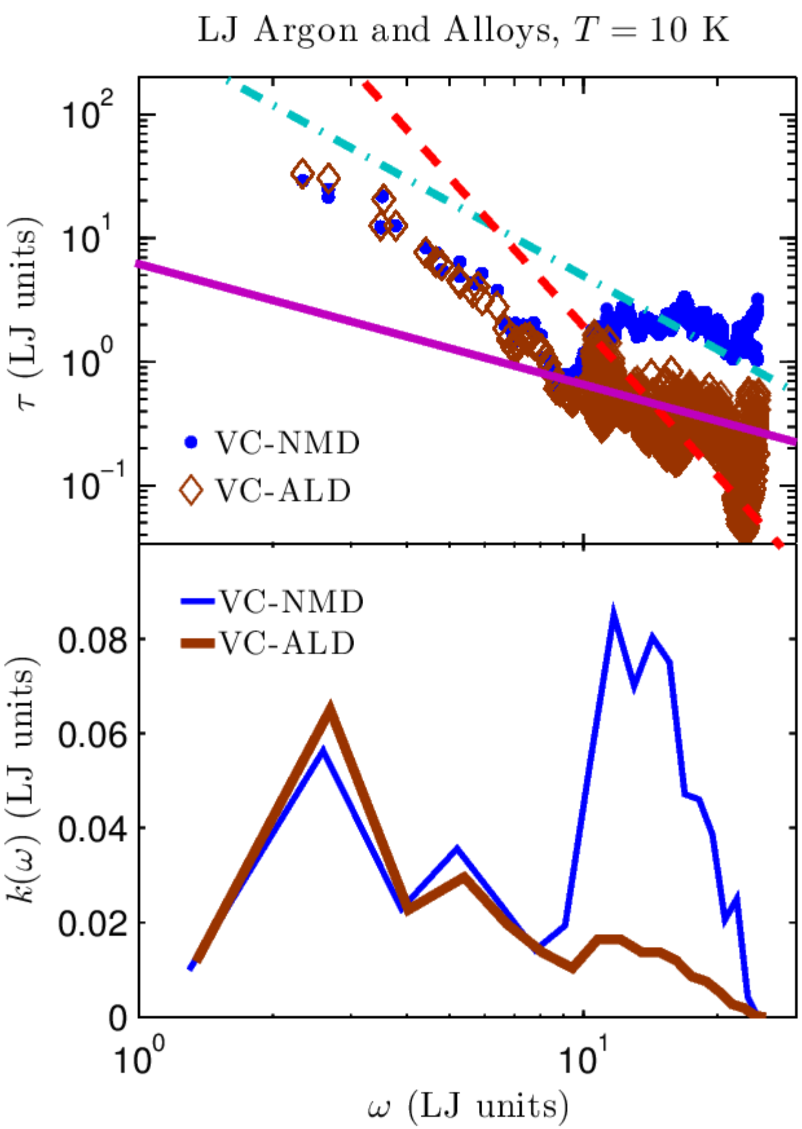
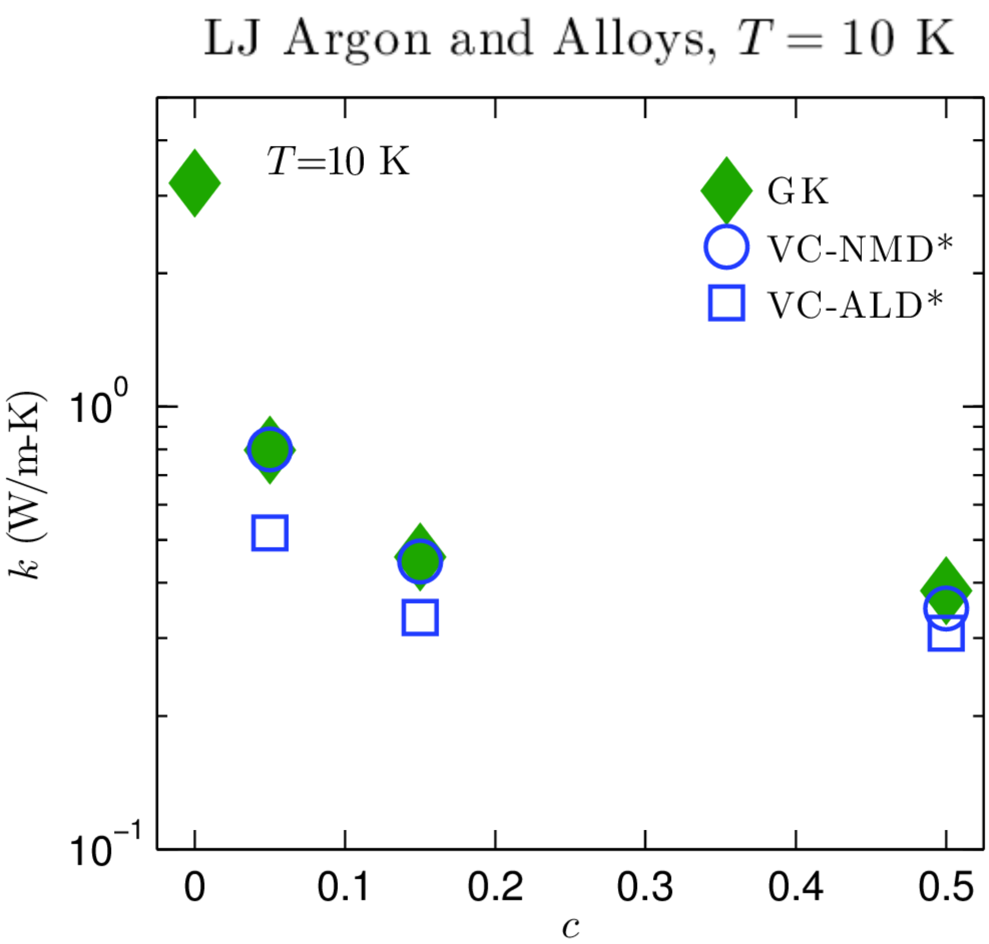
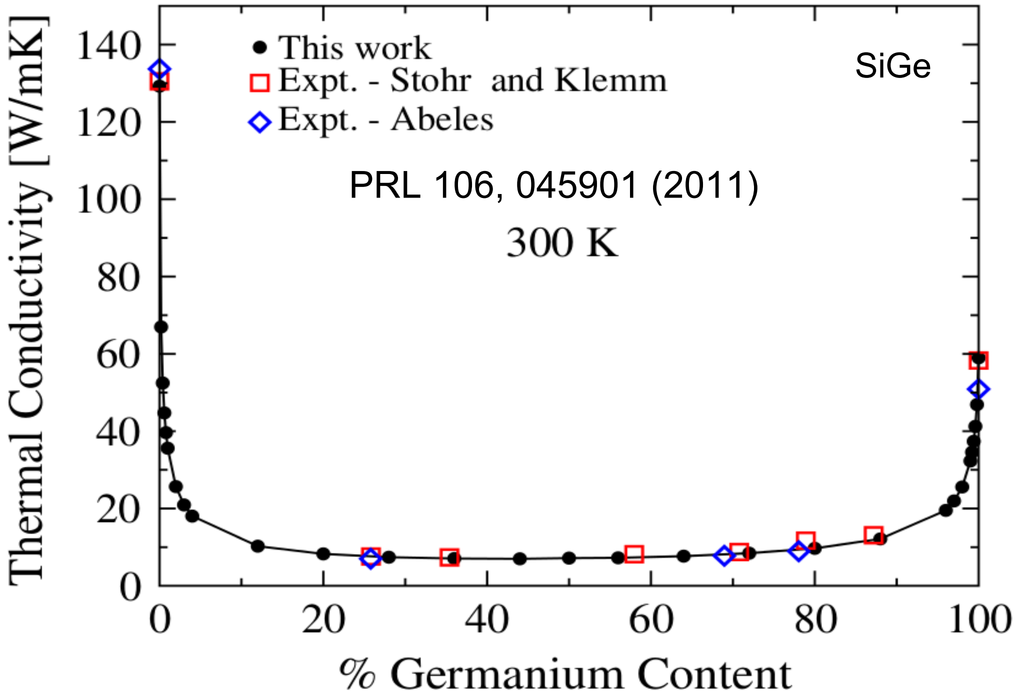


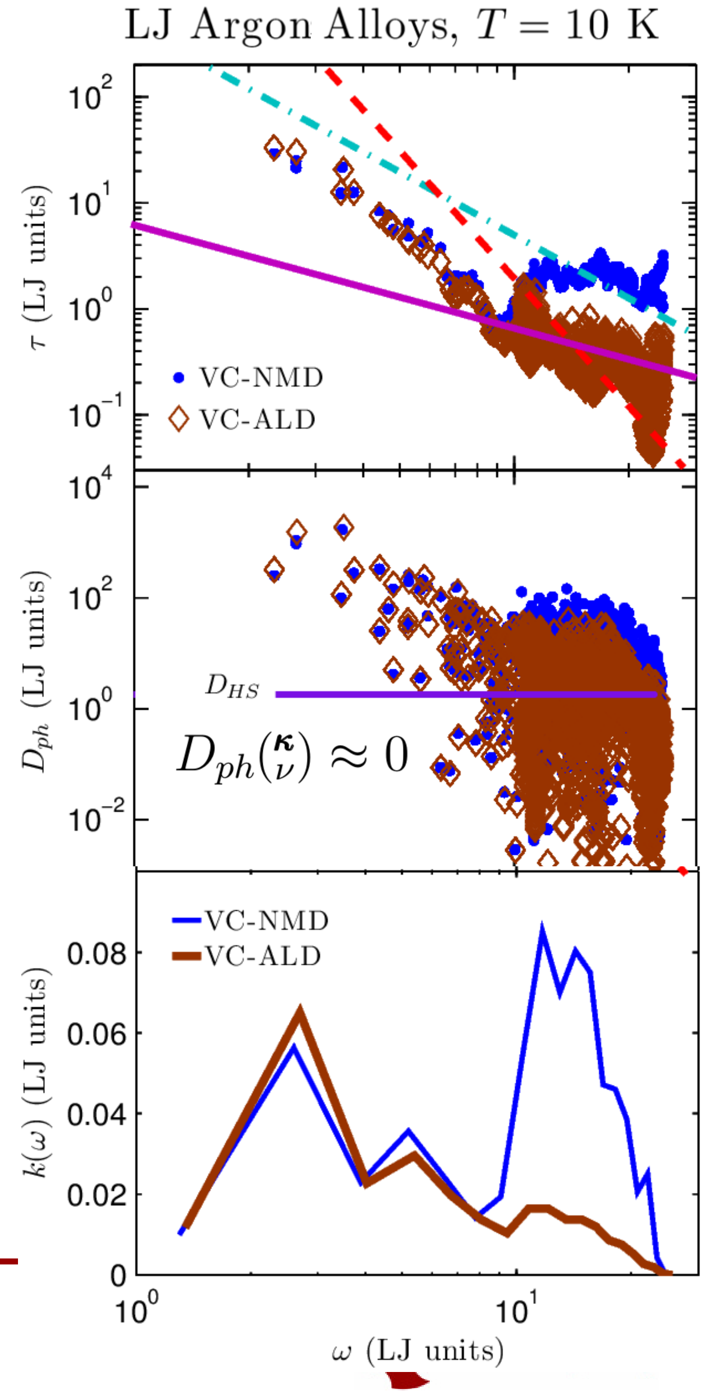
Thermal conductivity spectrum





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.

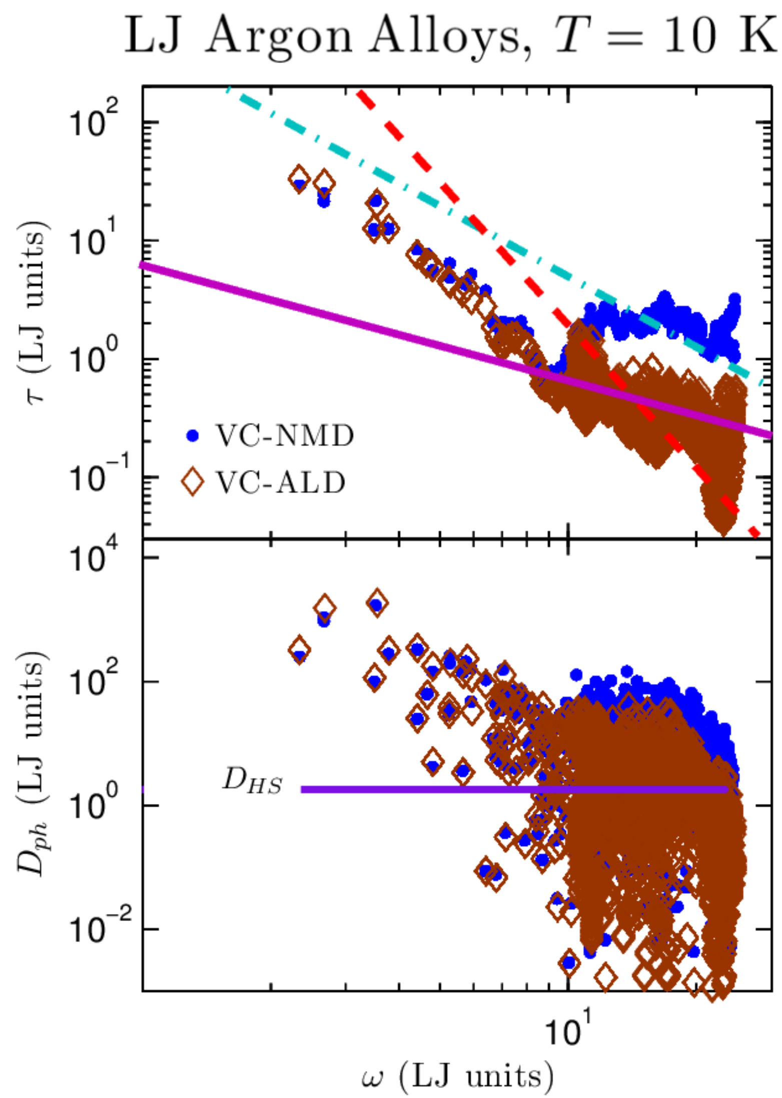
Summary

VC approximation underpredicts mode group velocities at high frequency.

$$D_{ph}\binom{\boldsymbol{\kappa}}{\nu} \approx 0 \qquad D_{HS} = \frac{1}{3} v_s a$$

VC-ALD underpredicts lifetimes at high-frequency.

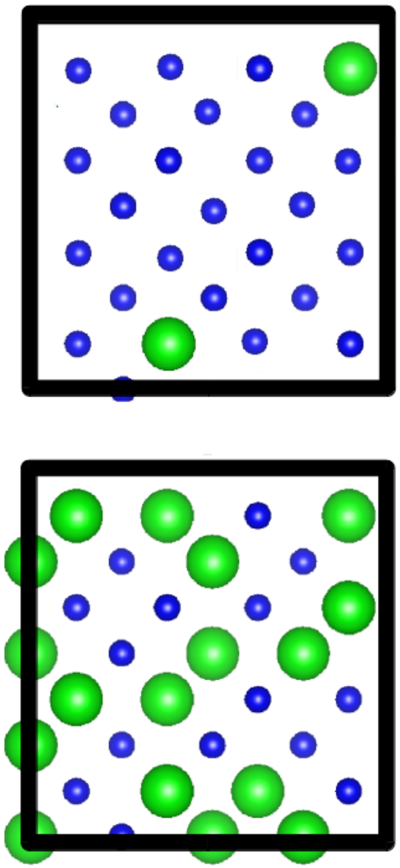
Breakdown of VC-ALD method is likely for materials near HS limit.



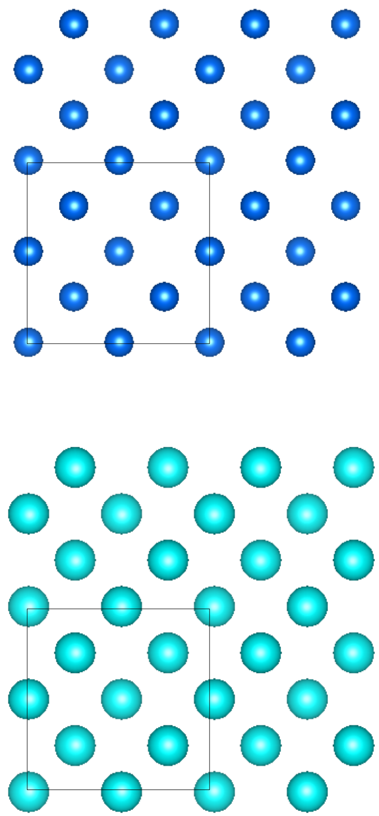
This work was supported by AFOSR award FA95501010098 and by a grant of computer time from the DOD High Performance Computing Modernization Program at the US Army Engineer Research and Development Center.

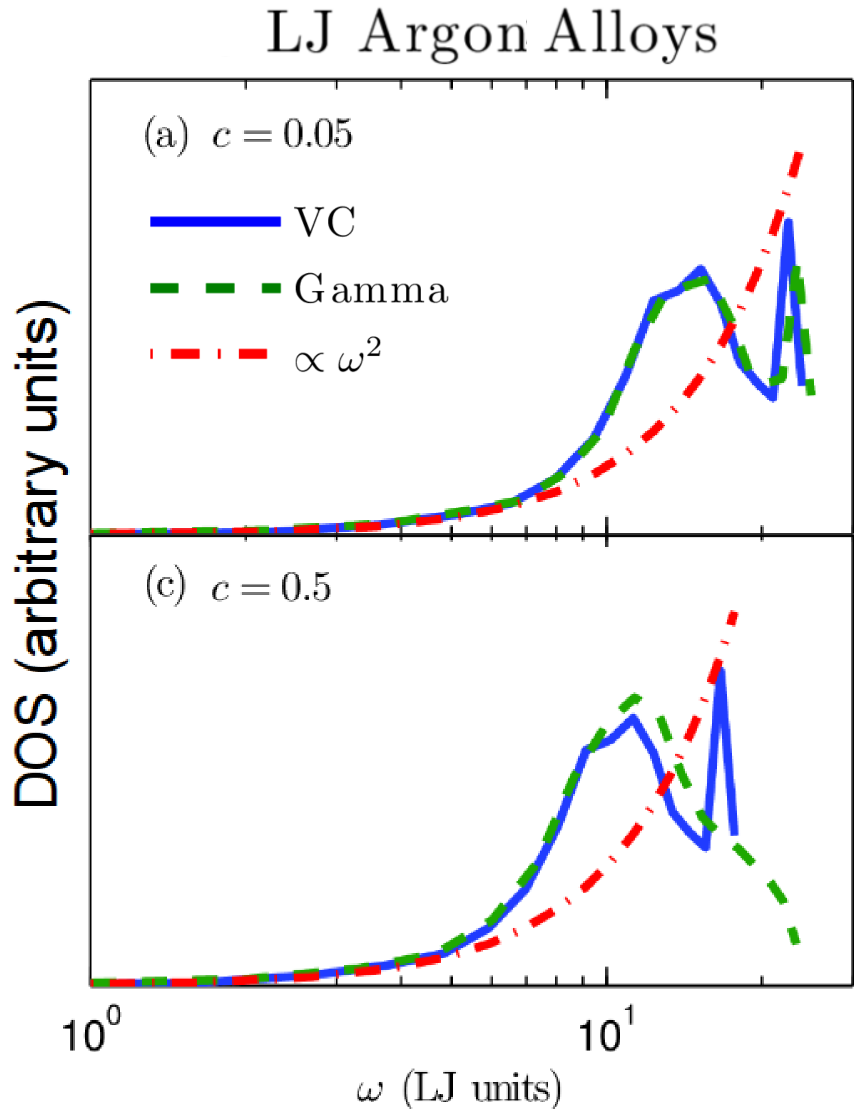
Explicit disorder: VC vs Gamma

Gamma

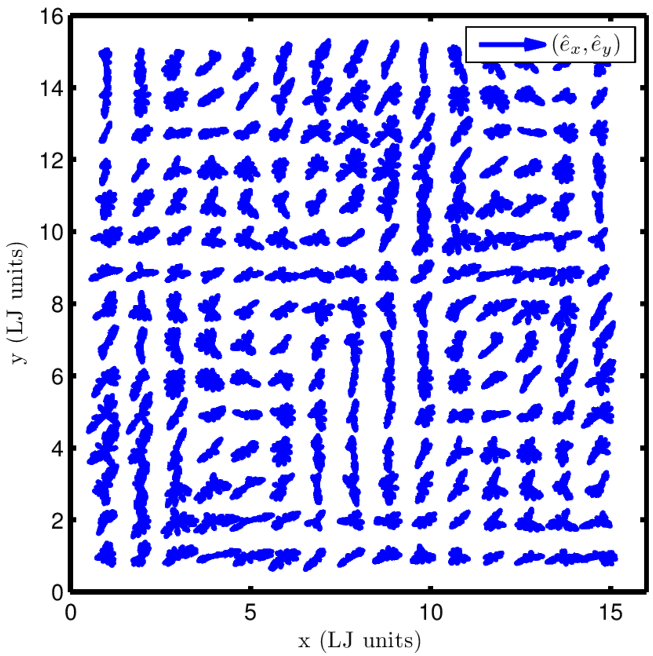


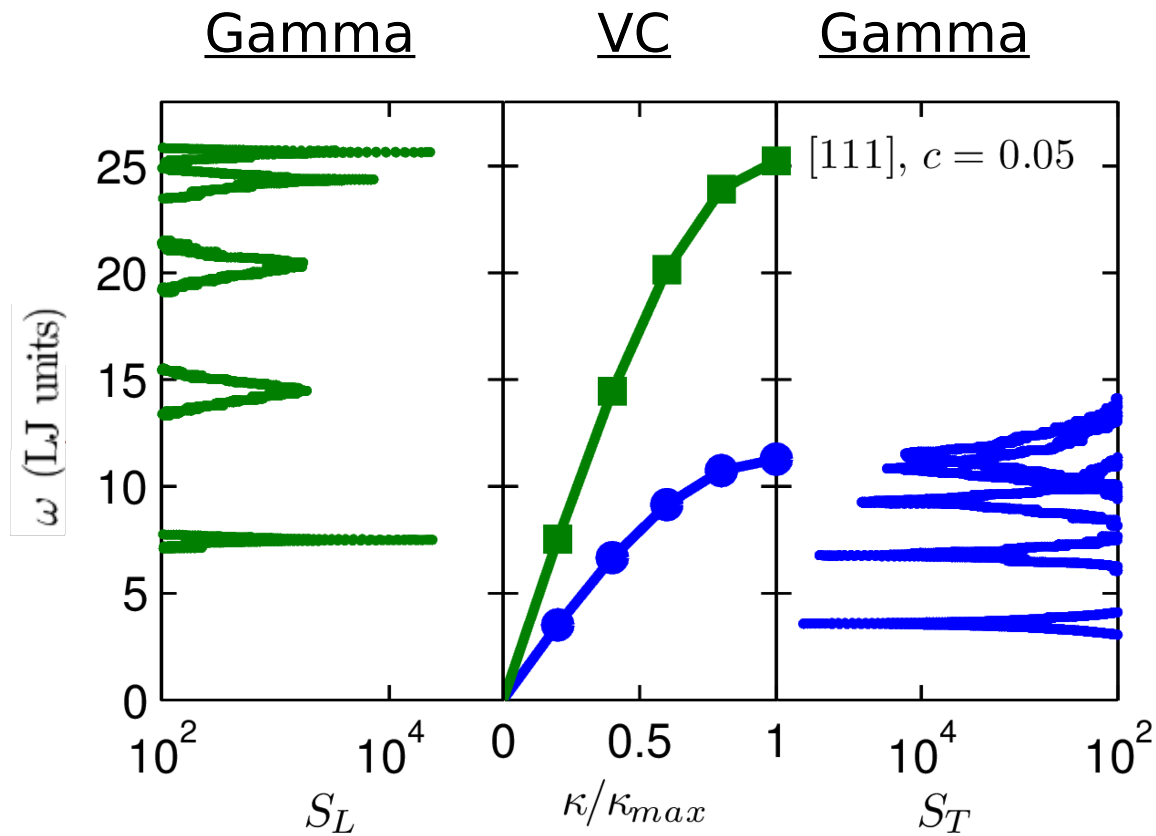
VC





Explicit disorder: Structure Factor

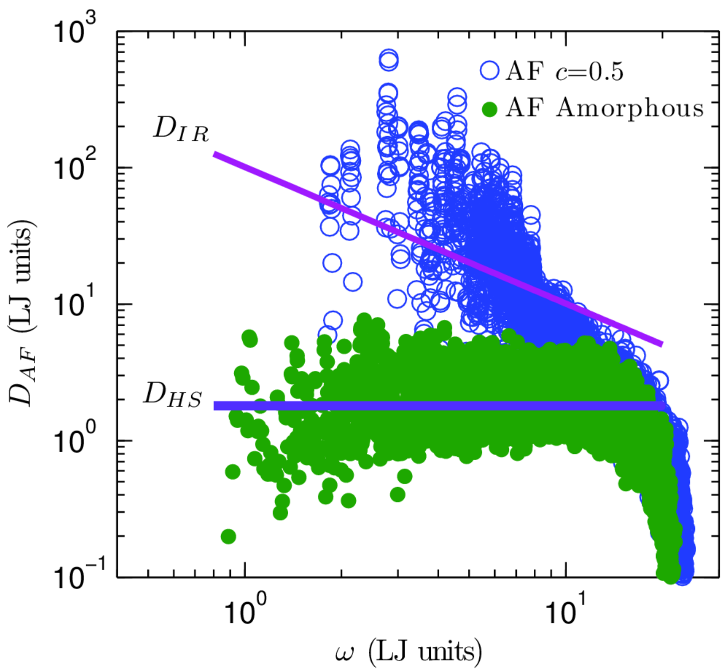


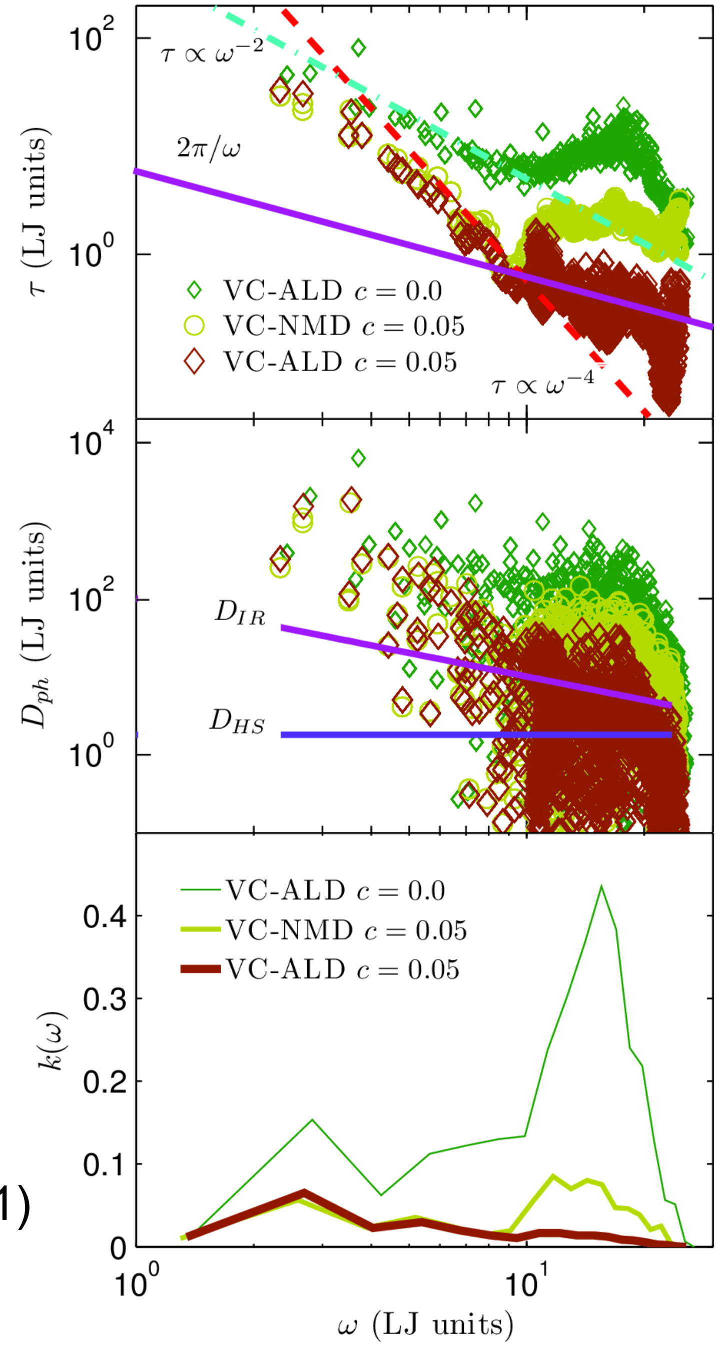


Phys. Rev. B 59, 35513559 (1999)

HS/IR Limit

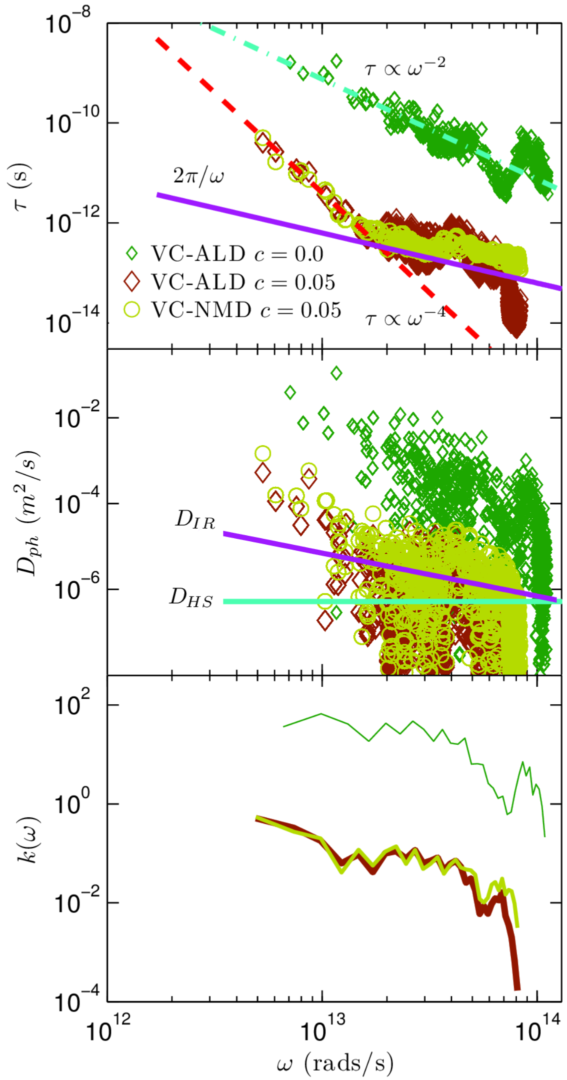
$$D_{IR} = \frac{2\pi}{3}\frac{v_s^2}{\omega}.$$

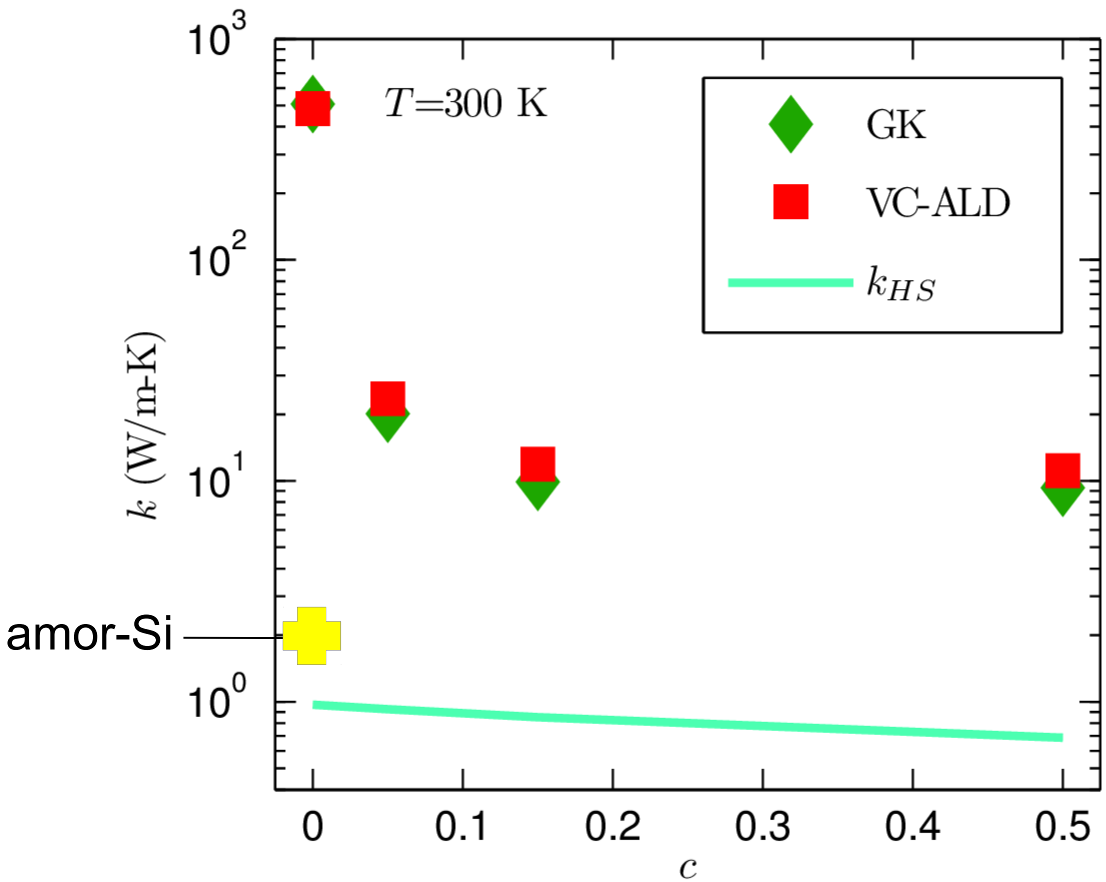




P. Sheng and M. Zhou, Science 253, 539542 (1991)

Thermal conductivity: SW silicon alloy

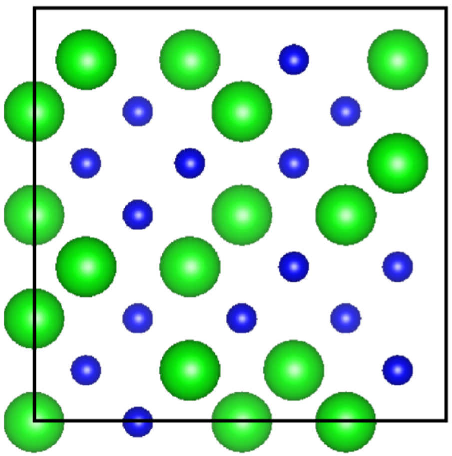




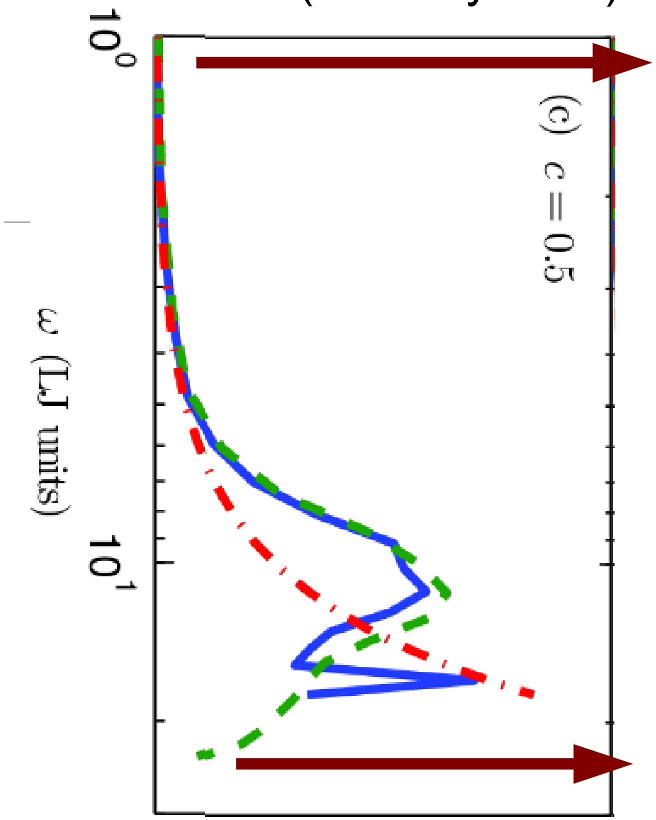
Gamma modes

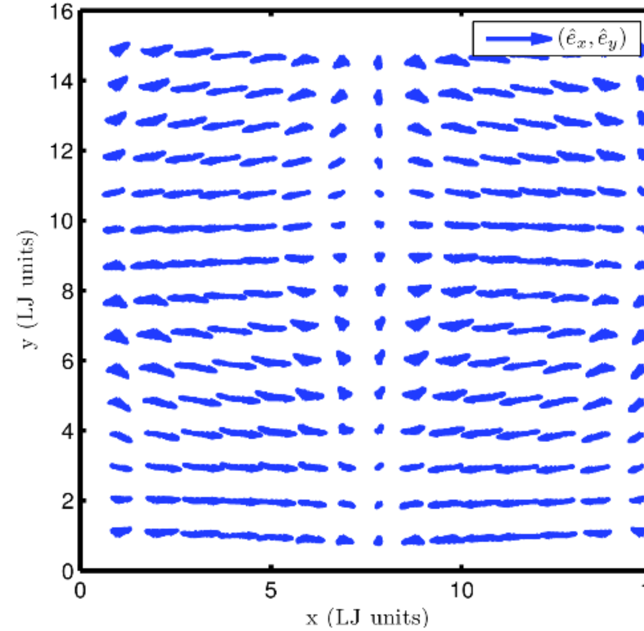
$$e\left(\begin{smallmatrix}\boldsymbol{\kappa}=0 & b\\ \nu & \alpha\end{smallmatrix}\right)$$

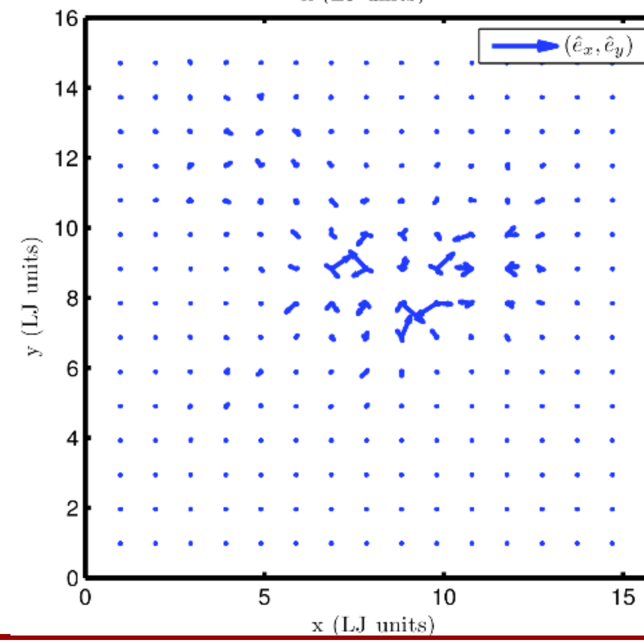
(a) disordered supercell



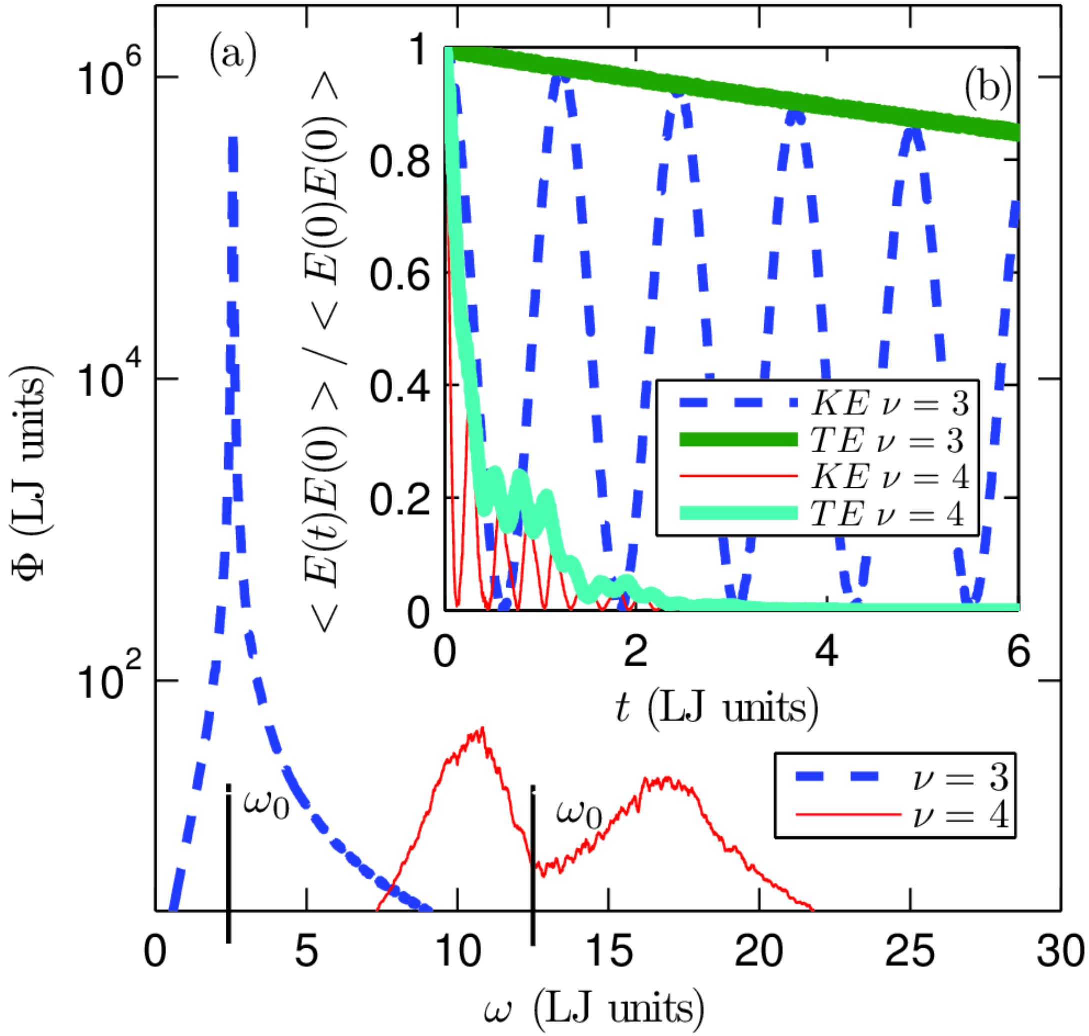
DOS (arbitrary units)



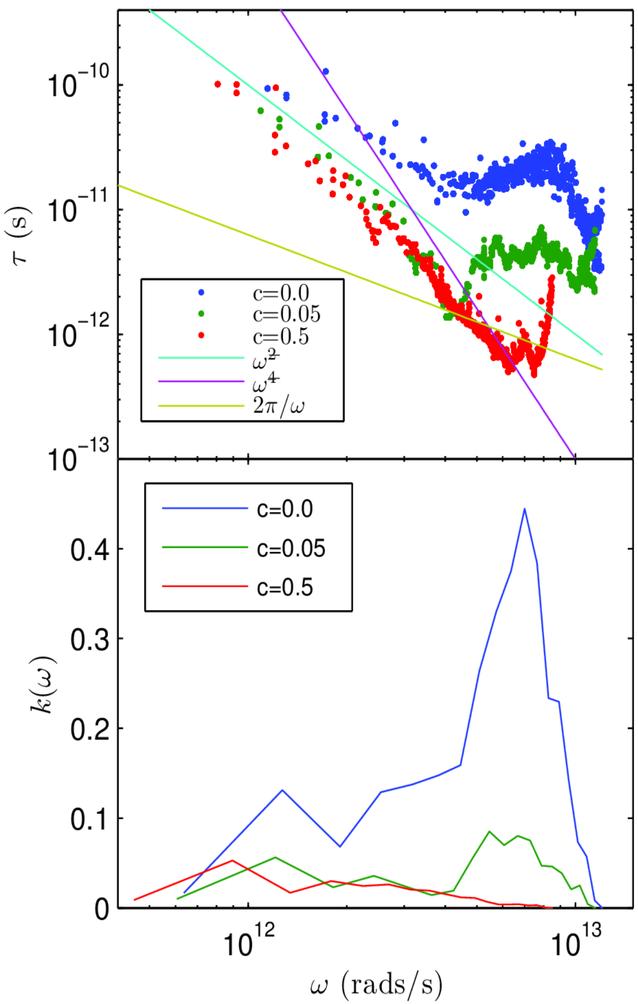


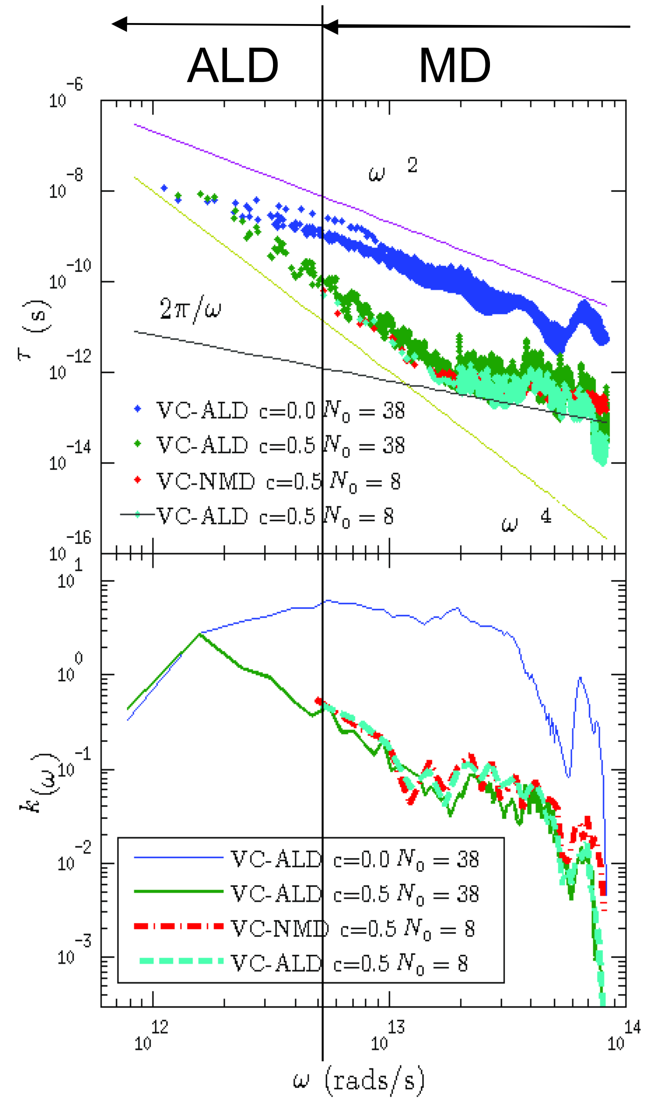


NMD using VC modes

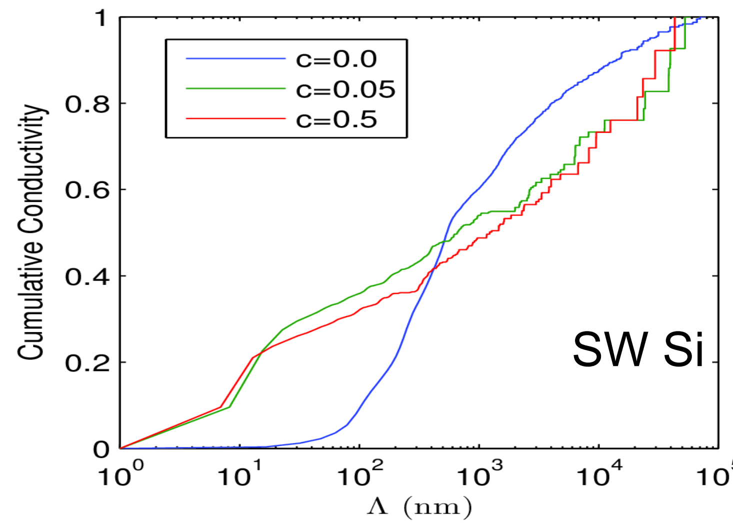


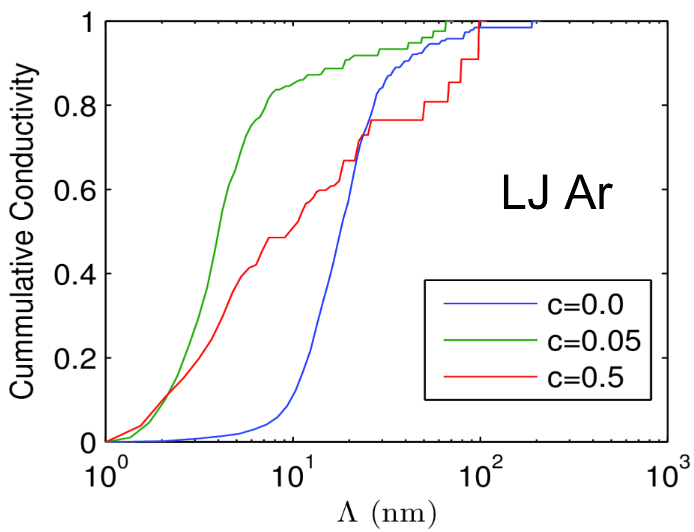
Phonon Spectrum: LJ Ar vs SW Si

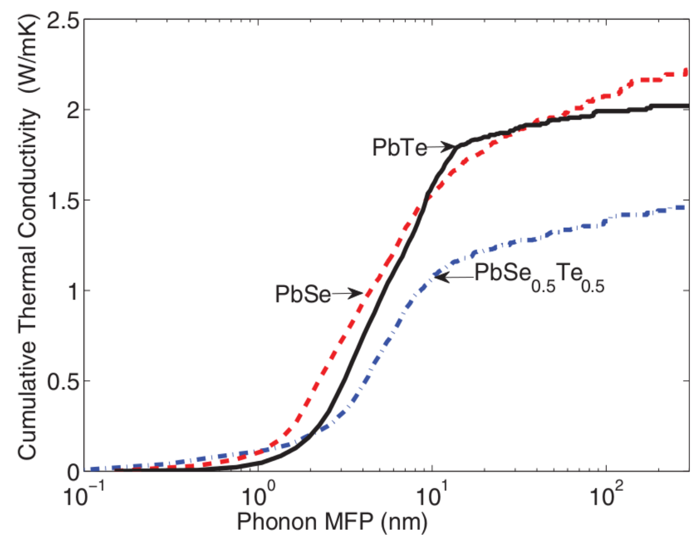




Conductivity Accumulation

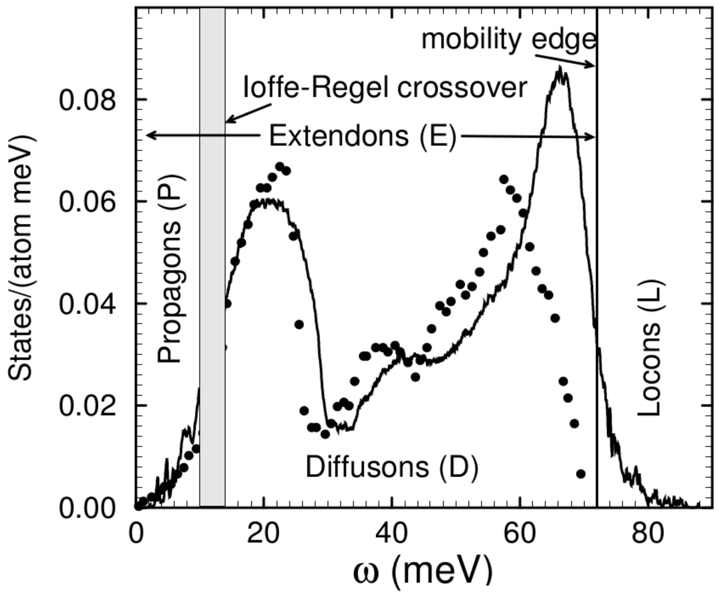


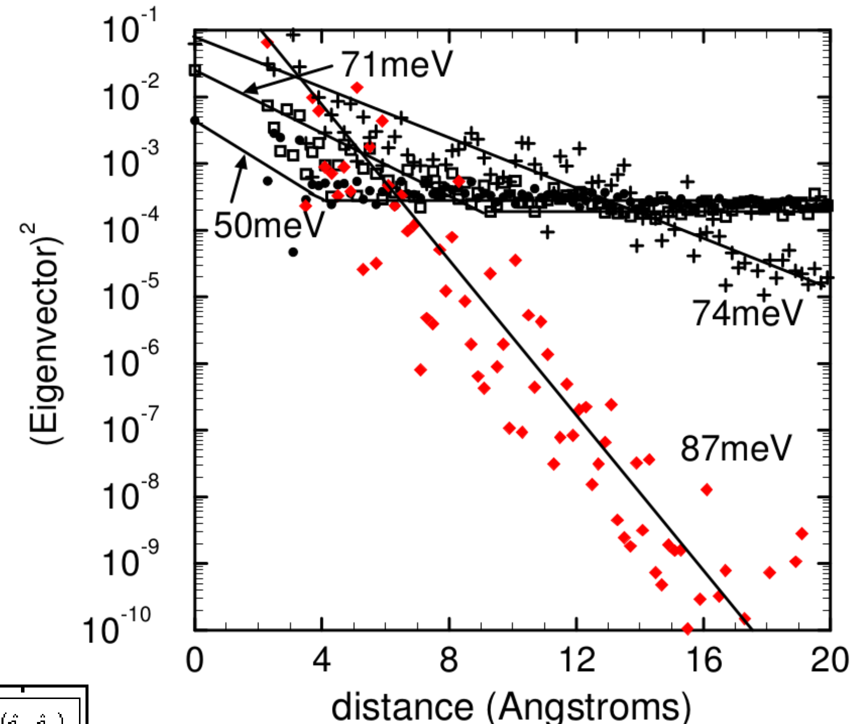


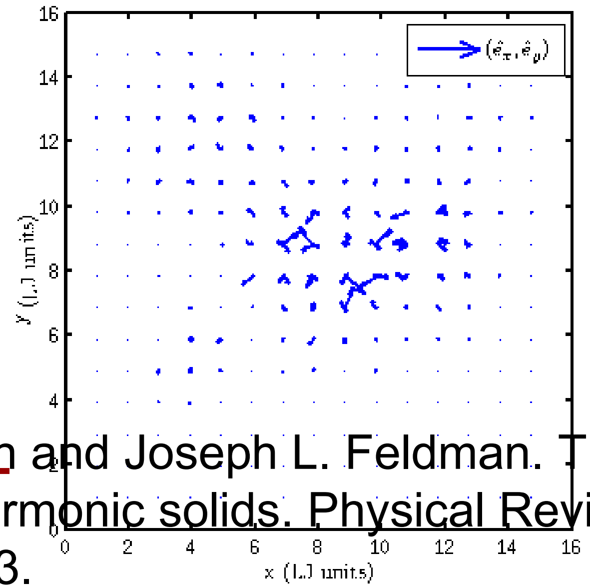


PHYSICAL REVIEW B 85, 184303 (2012)

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.