Evaluation of the Virtual Crystal Approximation for Predicting Thermal Conductivity

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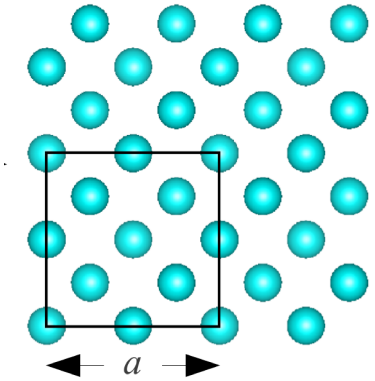
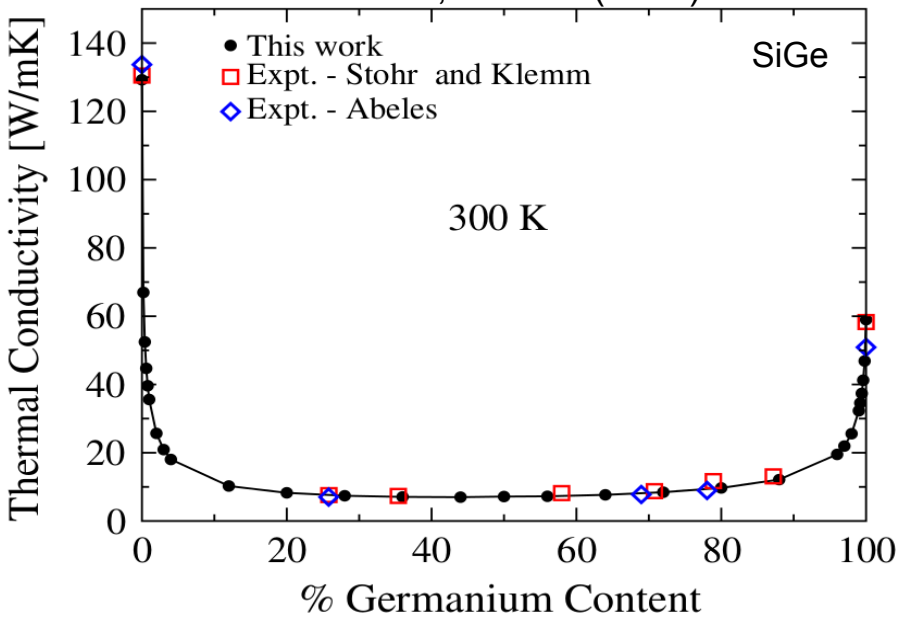
Motivation: experimental accuracy

Expensive Density Functional Theory (DFT) calculations
+
Virtual Crystal (VC) approximation &
Anharmonic Lattice Dynamics (ALD) (**VC-ALD**)

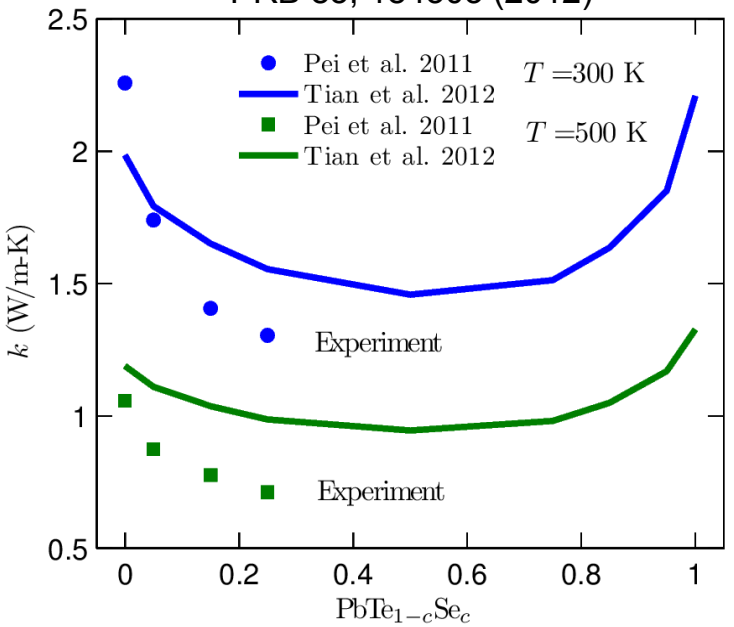


Alloys: isotopic effects, thermoelectric materials

PRL 106, 045901 (2011)



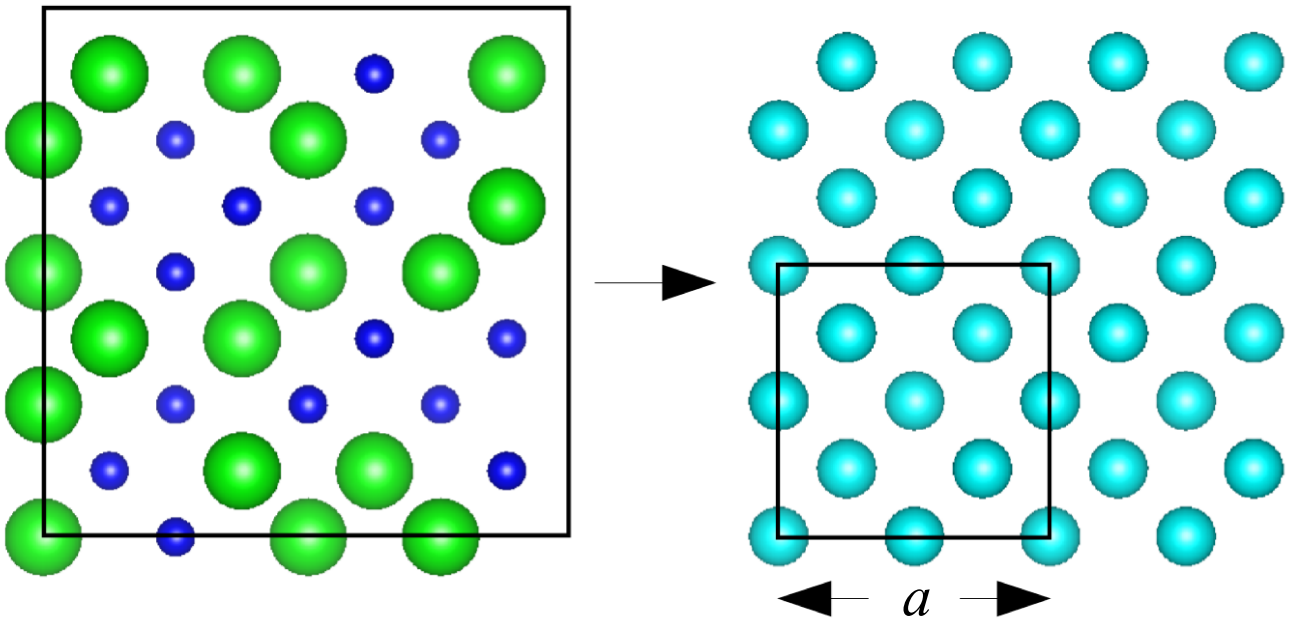
PRB 85, 184303 (2012)



Virtual Crystal Approximation

(a) disordered supercell

(b) VC unit cell



c^{μ}, m^{μ}

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

Kinetic Theory for Crystal

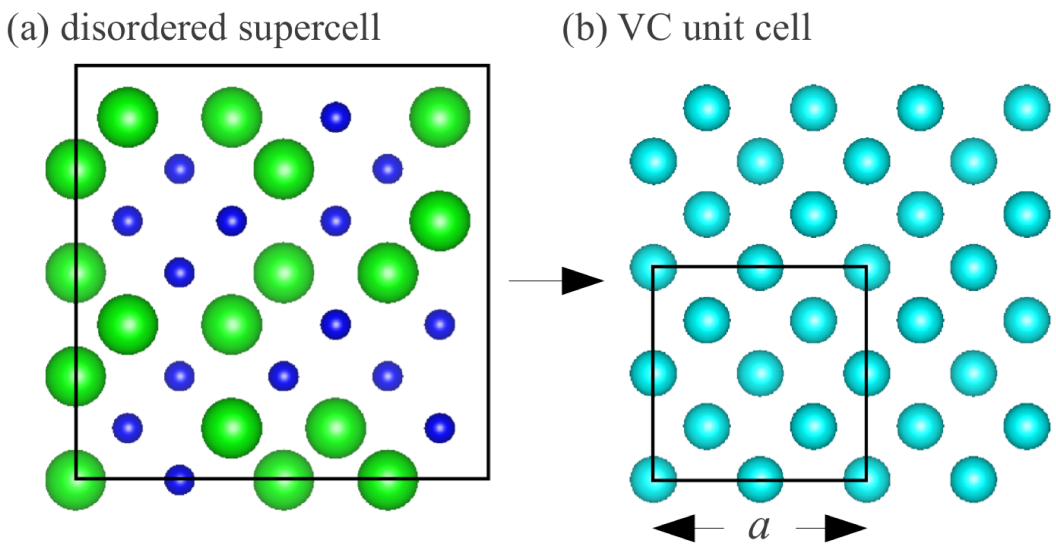
Thermal conductivity

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

Thermal diffusivity

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

VC-ALD: Lifetimes



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

ALD

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

Tamura

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

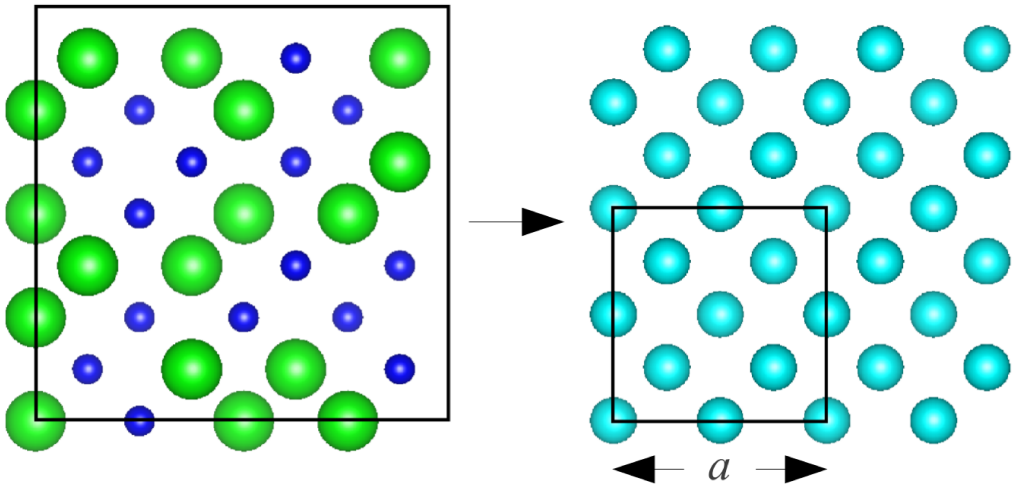
Matthiessen's Rule

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

VC-ALD: Group Velocity

(a) disordered supercell

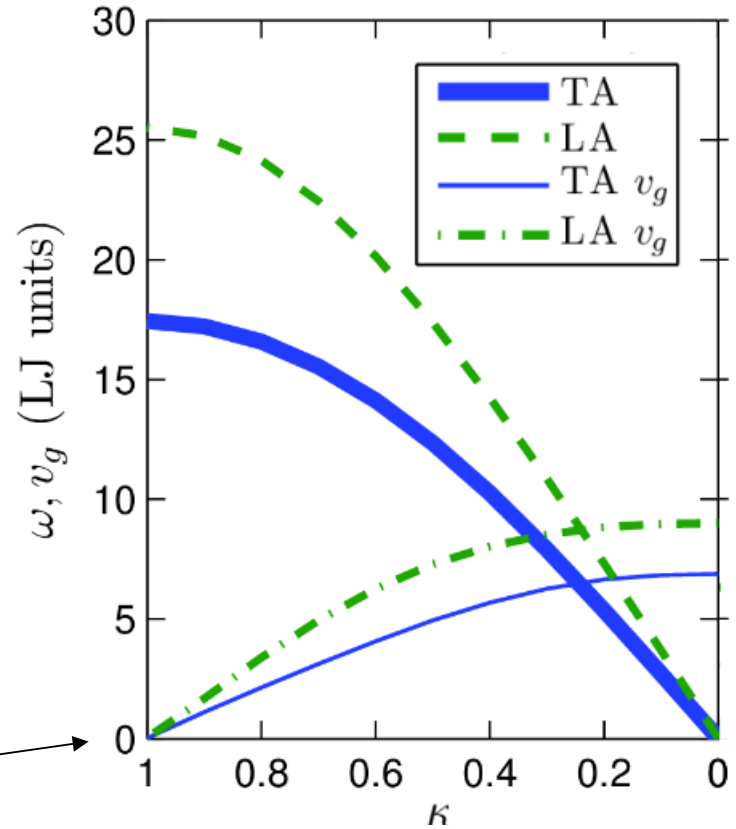
(b) VC unit cell



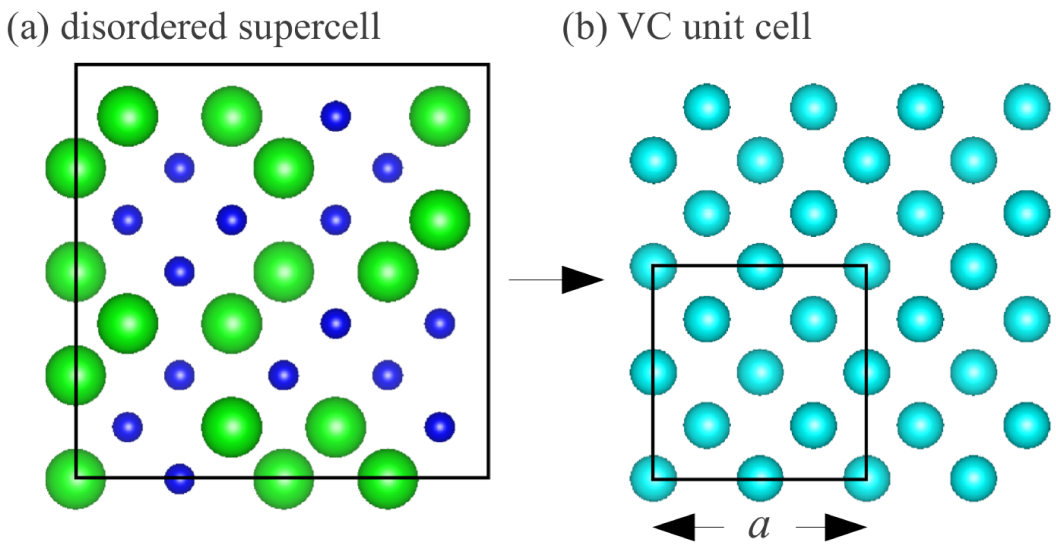
$$D_{ph,\mathbf{n}}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right) = v^2_{g,\mathbf{n}}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)\tau\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right)$$

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

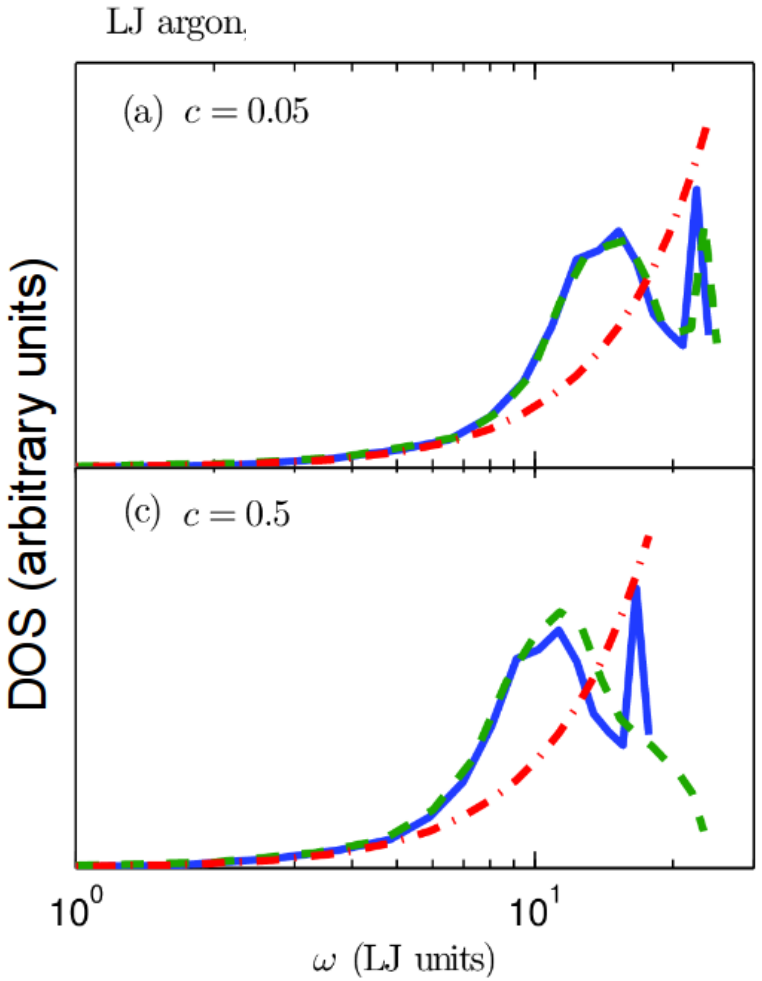
$$D_{ph}\left(\begin{matrix}\boldsymbol{\kappa}\\ \nu\end{matrix}\right) \approx 0$$



Explicit disorder: VC vs Gamma



Gamma

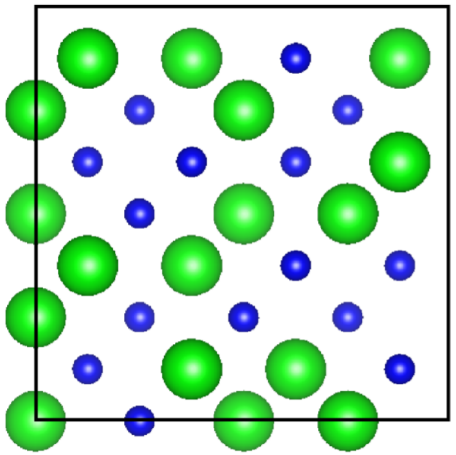


Computationally-inexpensive empirical potential:

Lennard-Jones argon

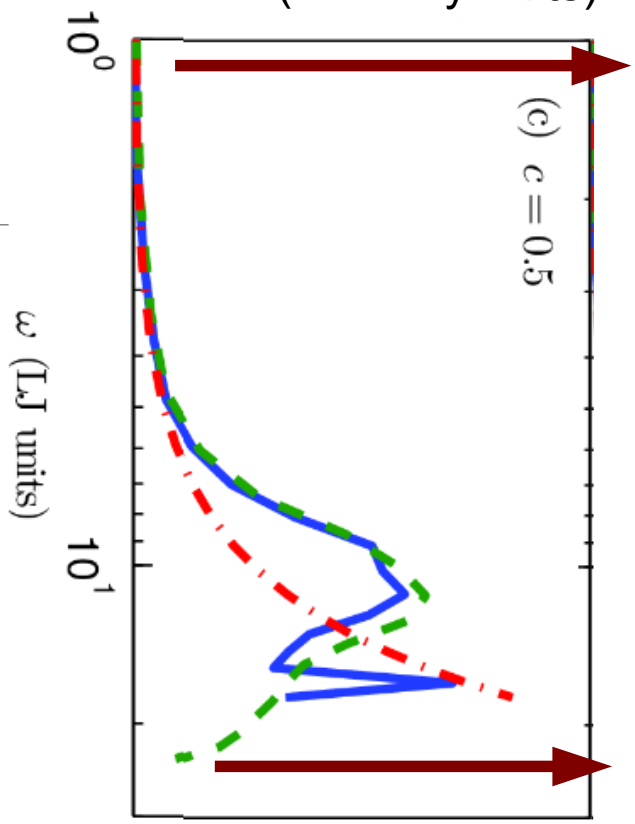
Gamma modes

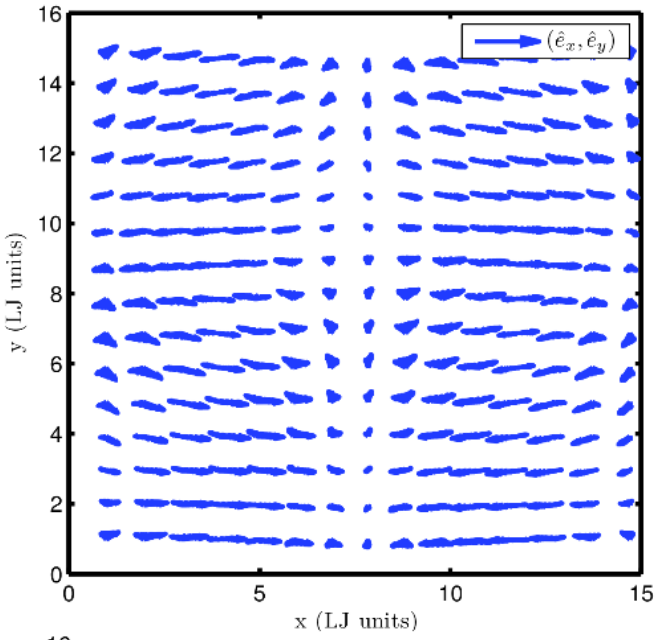
(a) disordered supercell

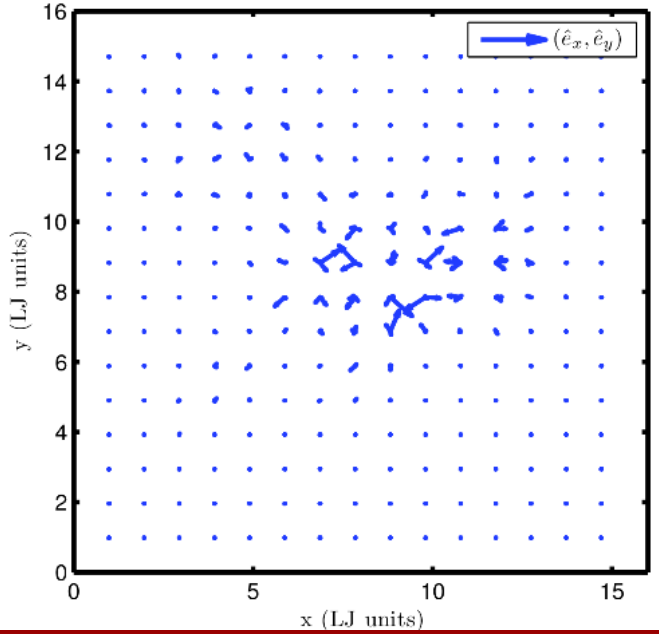


$e\begin{pmatrix}\boldsymbol{\kappa}=\mathbf{0} & b\\ \nu & \alpha\end{pmatrix}$

DOS (arbitrary units)



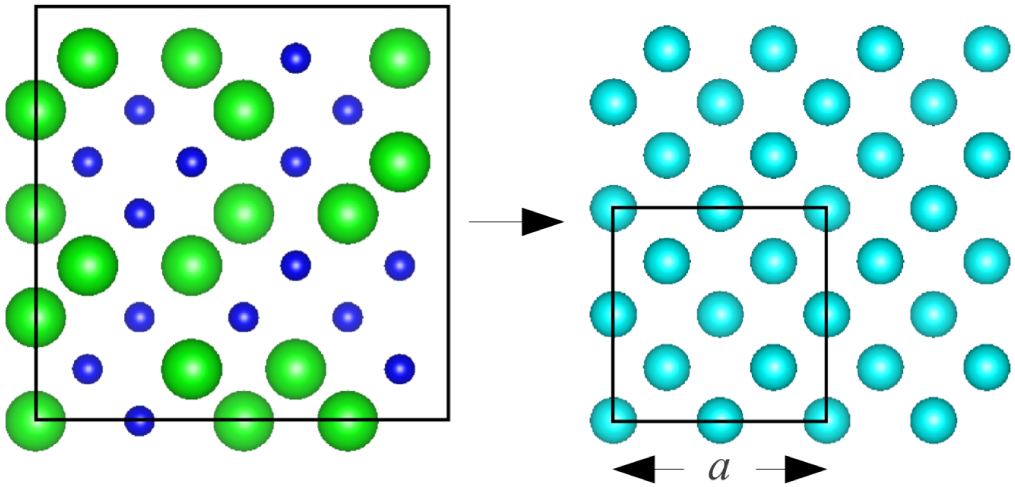




Gamma modes Structure Factor

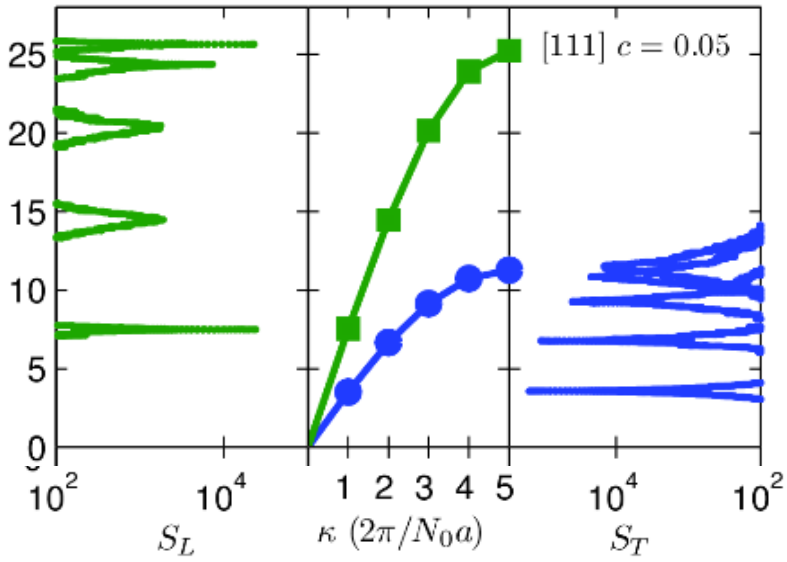
(a) disordered supercell

(b) VC unit cell



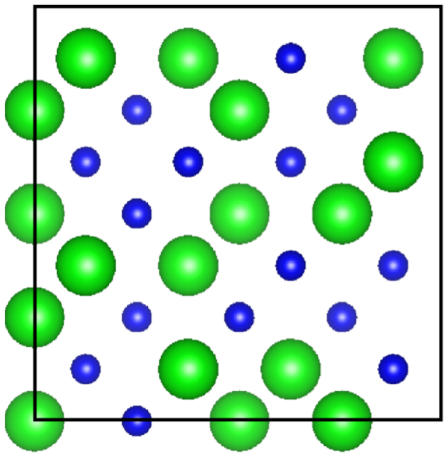
$$v_{g,\mathbf{n}}\binom{\boldsymbol{\kappa}}{\nu} = \frac{\partial \omega\binom{\boldsymbol{\kappa}}{\nu}}{\partial \boldsymbol{\kappa}}$$

Gamma VC Gamma



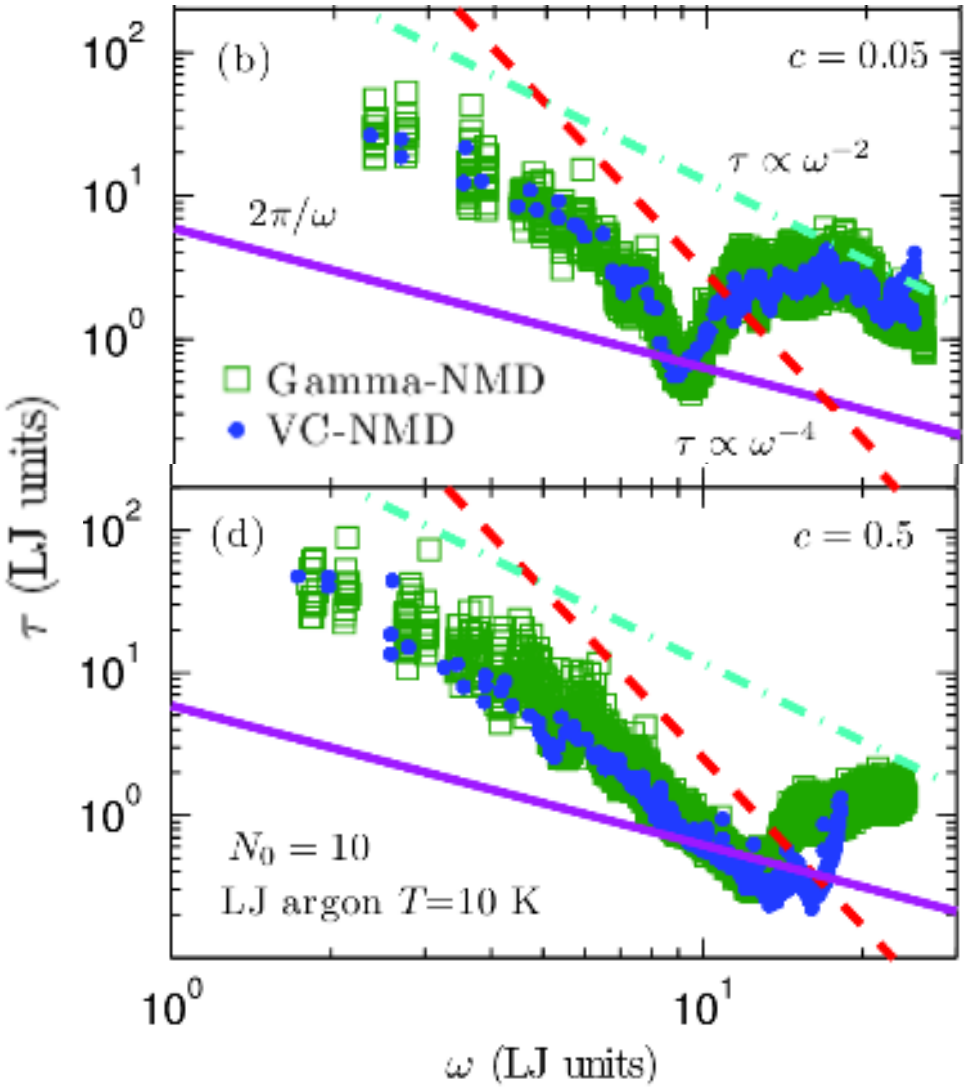
Normal Mode Decomposition (NMD)

(a) disordered supercell



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

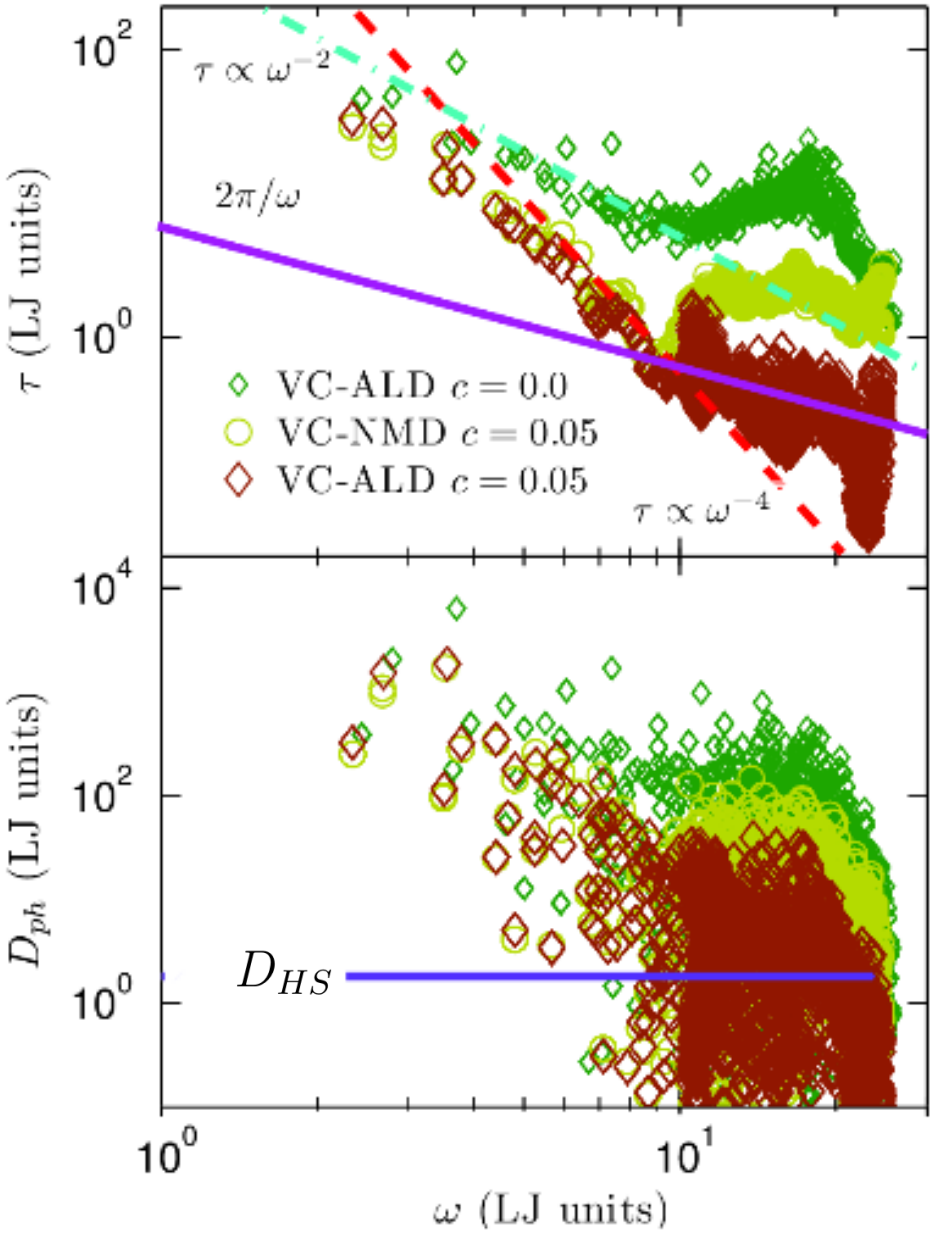
$$\tau(\kappa\,\nu) = \int_0^{t^*} \frac{< E(\kappa\,\nu;t)\, E(\kappa\,\nu;0) >}{< E(\kappa\,\nu;0)\, E(\kappa\,\nu;0) >} dt$$



VC Diffusivities

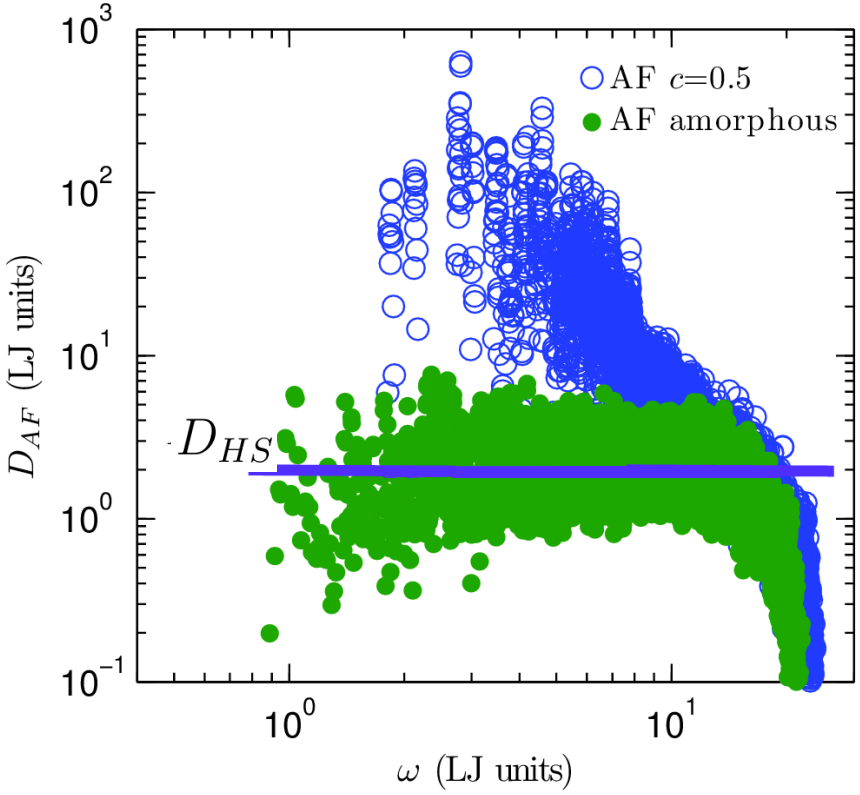
$$D_{ph,\mathbf{n}}(\kappa_\nu) = v^2_{g,\mathbf{n}}(\kappa_\nu)\,\tau(\kappa_\nu)$$

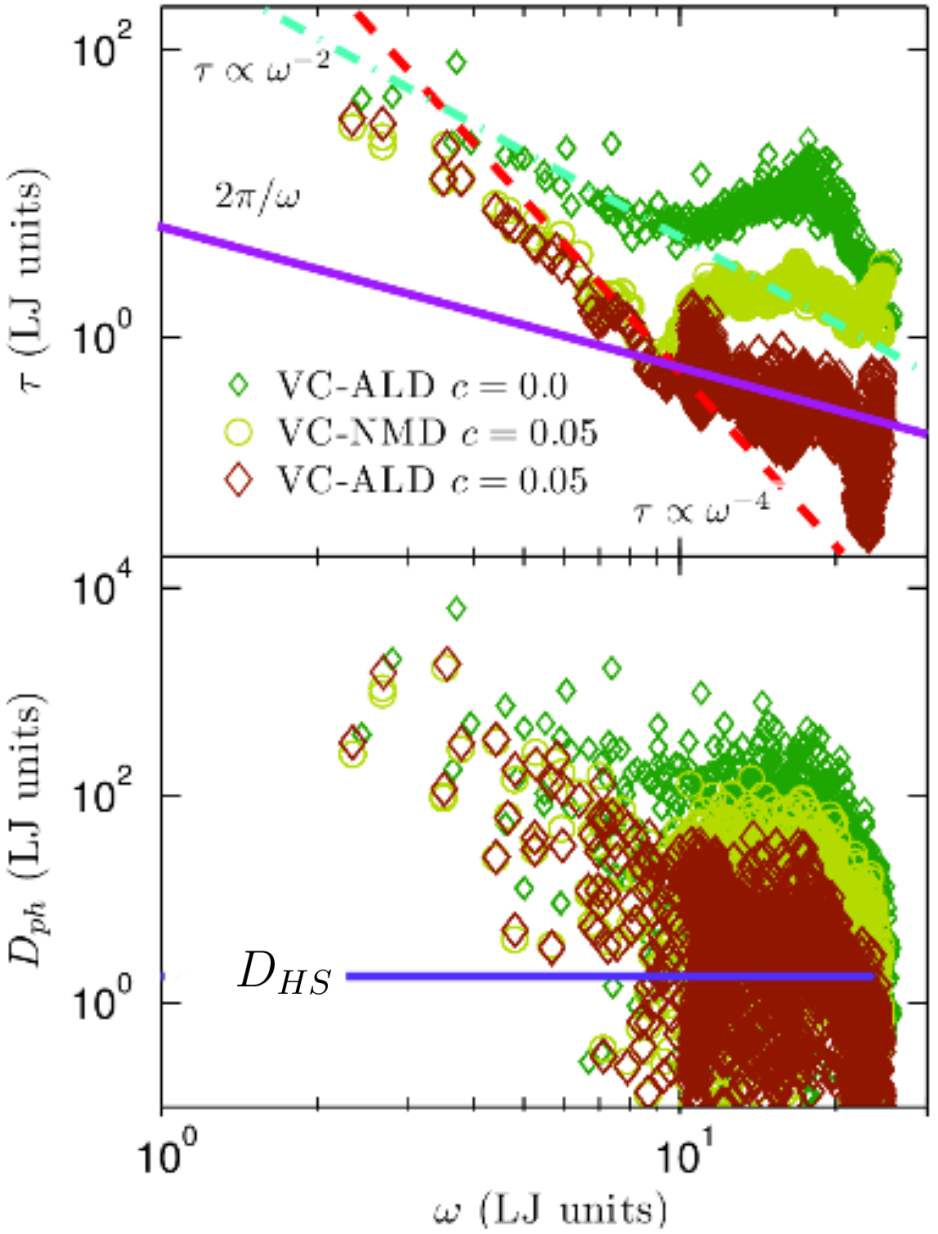
$$D_{HS} = \frac{1}{3} v_s a$$



AF Diffusivities

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





Thermal conductivity

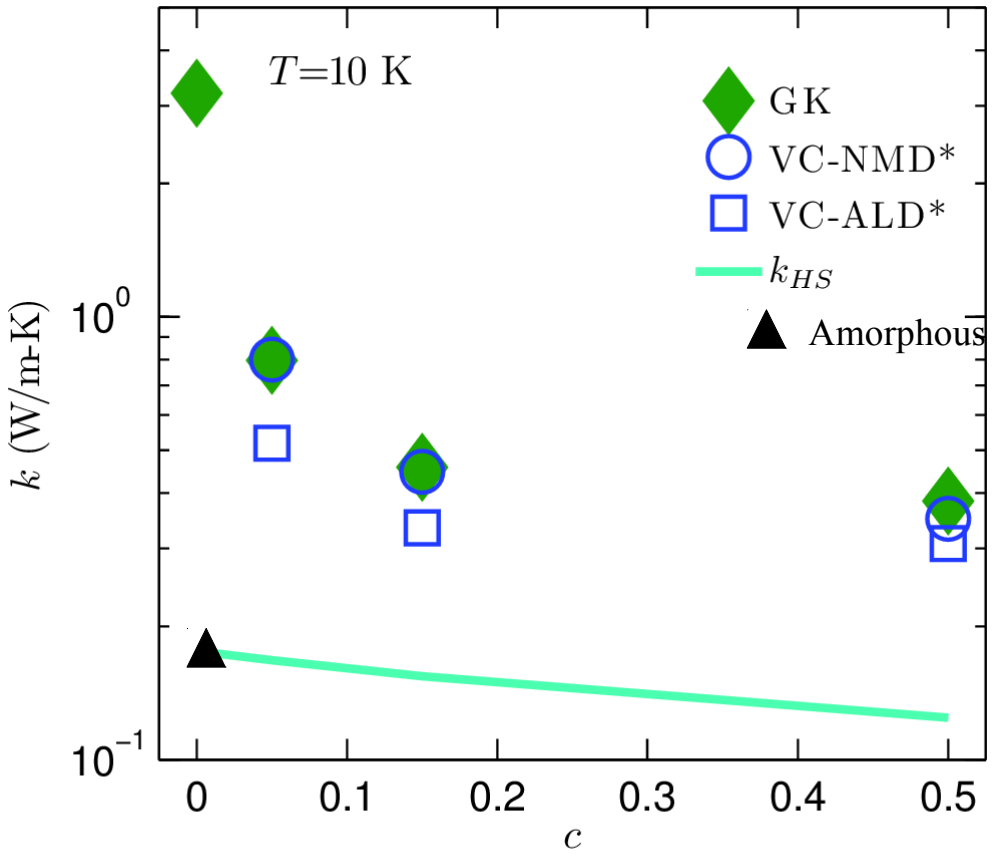
Green-Kubo (GK): top-down method

$$k_{HS} = \frac{k_B}{V_b} b v_s a$$

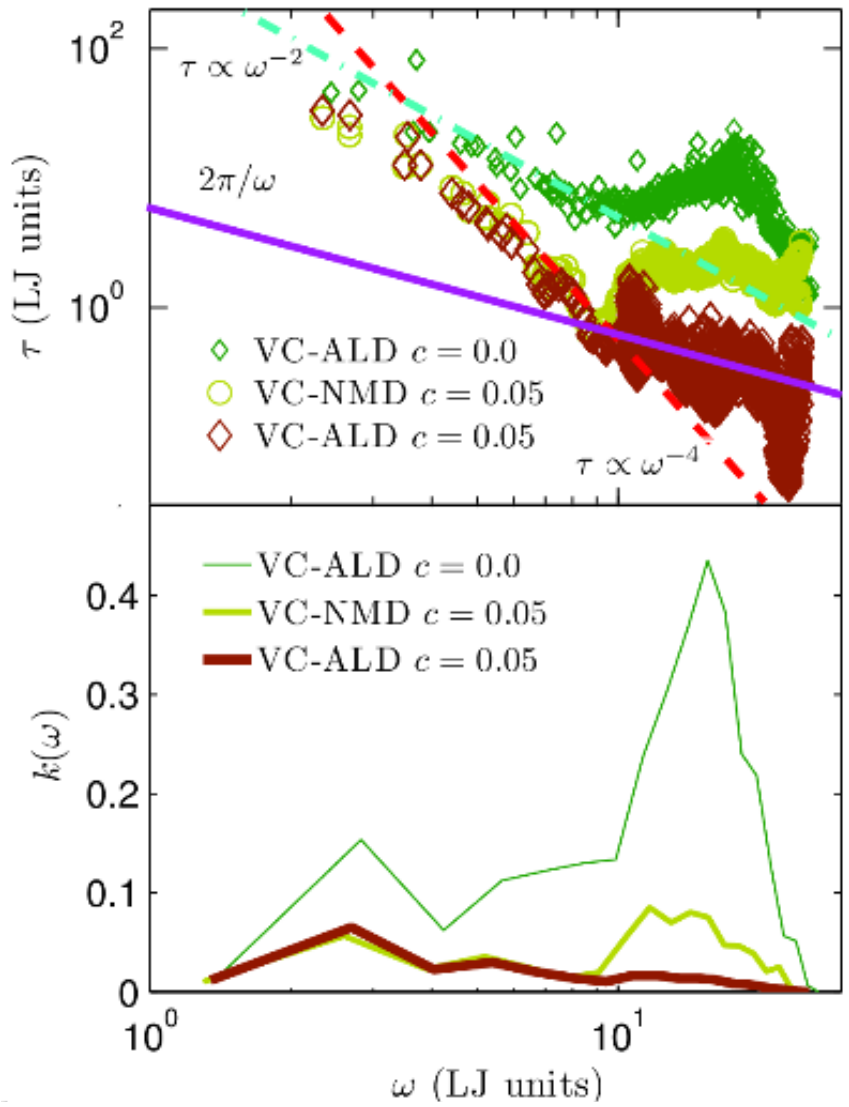
High-scatter adjustment*:

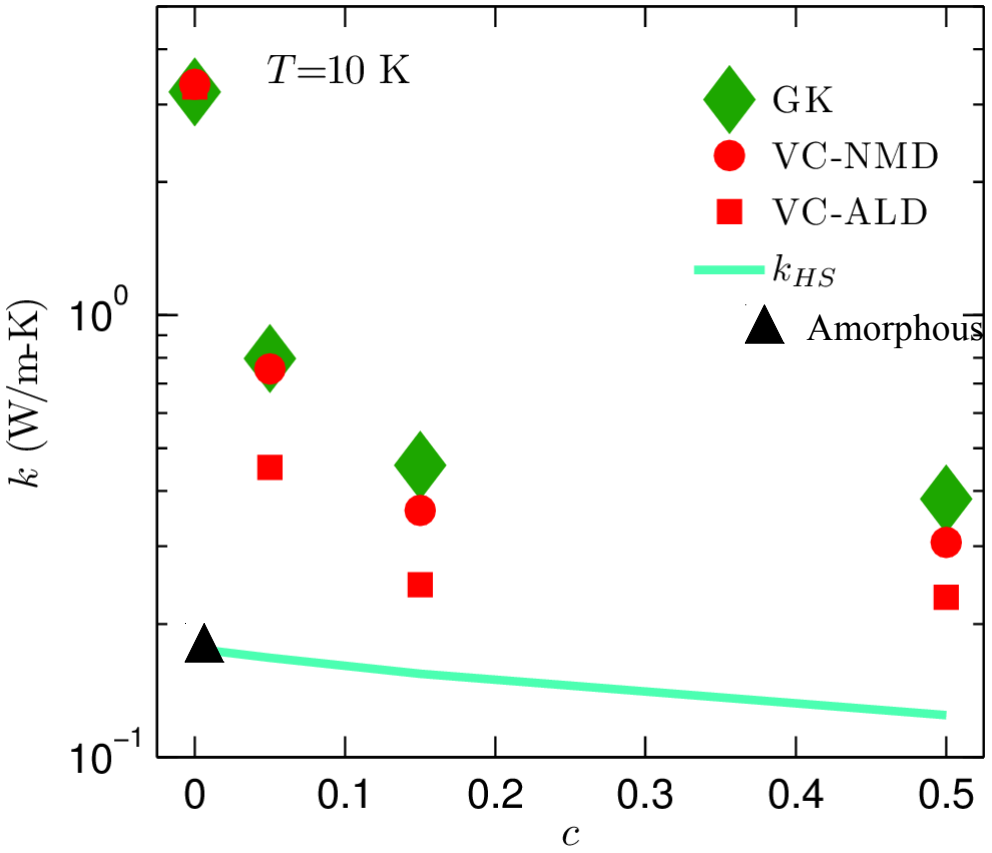
$$D_{ph}\left(\begin{matrix}\kappa\\ \nu\end{matrix}\right) \quad D_{HS}$$

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



Thermal conductivity spectrum





Summary