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# One-dimensional phonon effects in direct molecular dynamics method for thermal conductivity determination

L. Hu,<sup>1</sup> W. J. Evans,<sup>2</sup> and P. Keblinski<sup>1,a)</sup>

<sup>1</sup>*Department of Materials Science and Engineering, Rensselaer Polytechnic Institute, Troy, New York 12180, USA*

<sup>2</sup>*Rensselaer Nanotechnology Center, Rensselaer Polytechnic Institute, Troy, New York 12180, USA*

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# Phonon Modes

Sum over all modes

$$\kappa = \frac{1}{V k_B T^2} \sum_{k_x, k_y, k_z, p} v^2(\mathbf{k}, p) \tau(\mathbf{k}, p) c(\mathbf{k}, p),$$

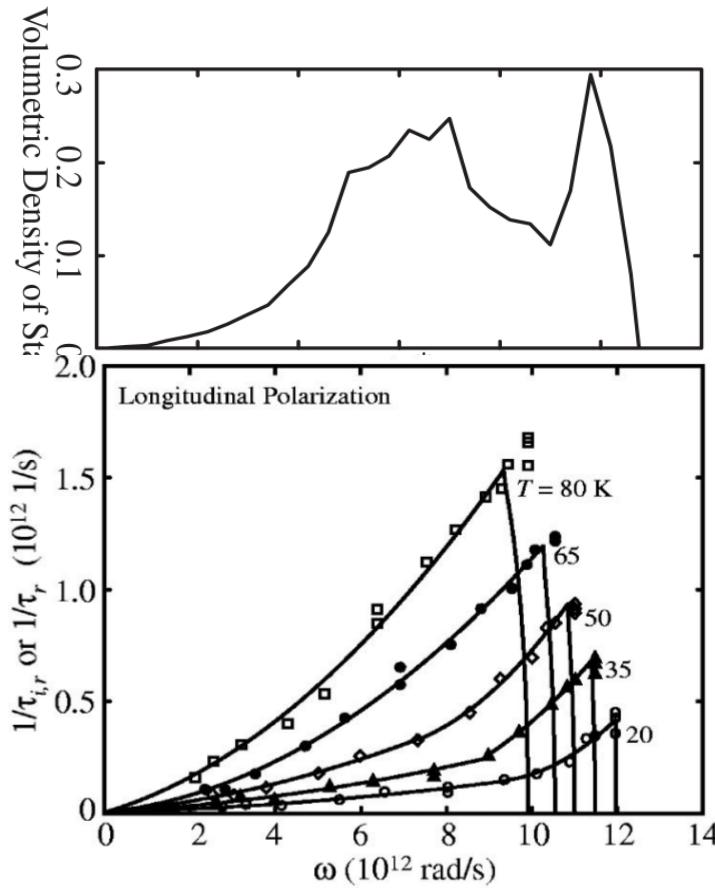
Continuum Limit

$$\kappa = k_B / 3V \int_0^{\omega_{\max}} v(\omega)^2 \tau(\omega) D(\omega) d\omega.$$

# 3D Isotropic Crystal

Debye approximation

$$\kappa = C_1 k_B / 3V \int_0^{\omega_D} \frac{1}{\omega^2} v^2 D(\omega) d\omega,$$



$$D(\omega) = \frac{V\omega^2}{2\pi^2 v^3}. \quad \text{DOS}$$

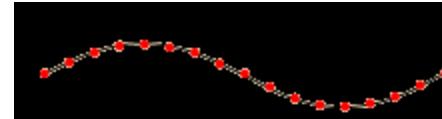
$$\tau(\omega) = C_1 / \omega^2 \quad \text{Umklapp scattering}$$

$$\kappa = \frac{C_1 k_B \omega_D}{6\pi^2 v}$$

# 1D Crystal

1D DOS is divergent with length

$$D(\omega) = \frac{L}{2\pi\nu},$$



Umklapp scattering?

$$\tau(\omega) = C_1/\omega^2$$

1D DOS is divergent with length = thermal conductivity divergent

$$\kappa \sim \int_{2\pi\nu/L}^{\omega_D} \frac{1}{\omega^2} d\omega \sim L.$$

# 3D Isotropic Crystal with aspect ratio 5

Sum over allowed wavevectors

$$\kappa = \frac{k_B}{3V} \sum_{k_x, k_y, k_z} v^2 \tau(\mathbf{k}),$$

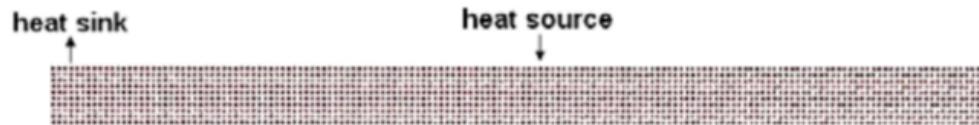
Debye approximation and Umklapp scattering

$$\tau(\omega) = C_1 / \omega^2$$

System with “aspect ratio”

$$\kappa = \frac{C_1 k_B}{3A\pi} \sum_{k_x} \sum_{k_y} \int_{2\pi/L_z}^{\pi/a} \frac{1}{k_x^2 + k_y^2 + k_z^2} dk_z,$$

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# NEMD

Simulation with high aspect ratio

Thermal conductivity from Fourier Law

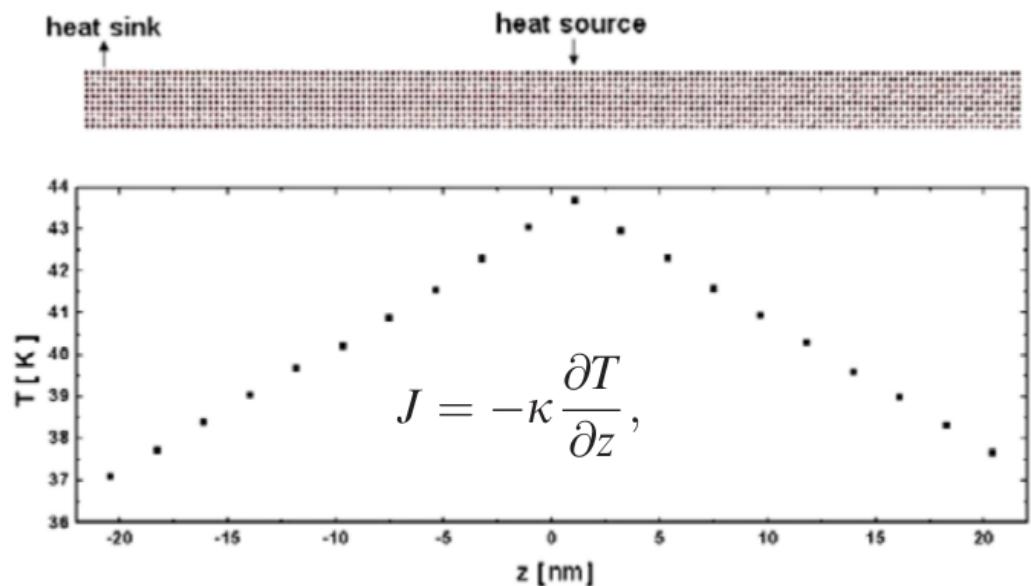


FIG. 2. (Color online) Typical temperature profile for periodic simulation cell of length  $L_Z$  and cross-section area  $A$ . Energy  $\Delta\varepsilon$  is added at source location each MD time step. Similarly, the same amount of energy is removed from a sink location each MD step. After a sufficient number of MD steps, a steady state thermal current and resulting temperature profile develops.

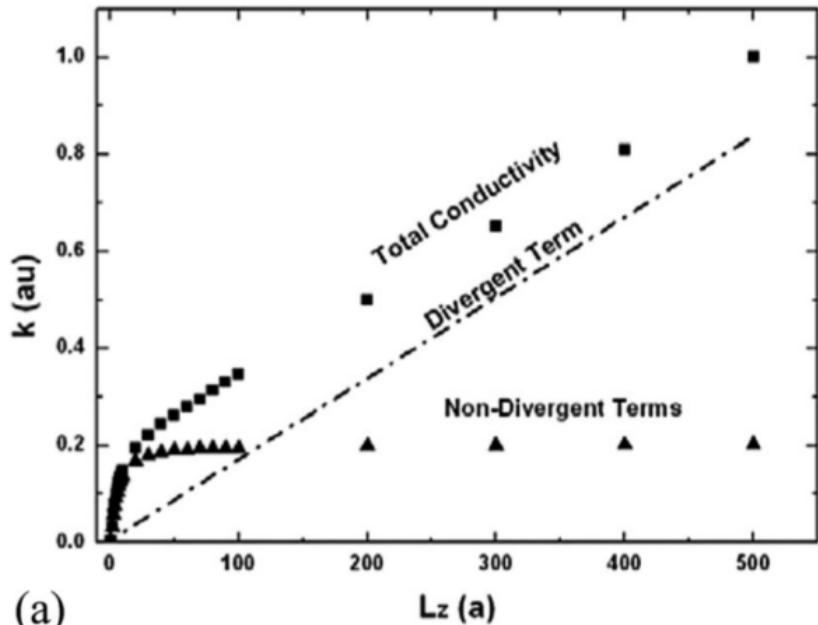
# NEMD results

$$\kappa = \frac{C_1 k_B}{3A\pi} \sum_{k_x} \sum_{k_y} \int_{2\pi/L_z}^{\pi/a} \frac{1}{k_x^2 + k_y^2 + k_z^2} dk_z,$$

$$\kappa = \frac{C_1 k_B}{3A\pi} \sum_{k_x} \sum_{k_y} \tan^{-1} \left( k_z \left/ \left( k_x^2 + k_y^2 \right)^{1/2} \right. \right) \left/ \left( k_x^2 + k_y^2 \right)^{1/2} \right|_{\pi/L_z}^{\pi/a}.$$

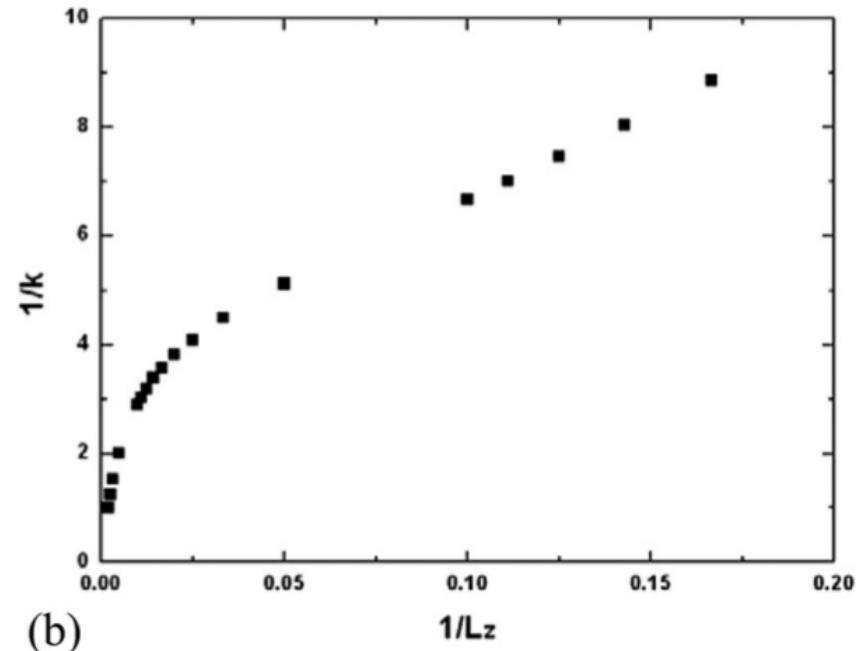
$$\kappa(k_x = k_y = 0) = \frac{C_1 k_B}{3A\pi^2} L_z.$$

## Non-Divergent Terms



(a)

## Divergent Term



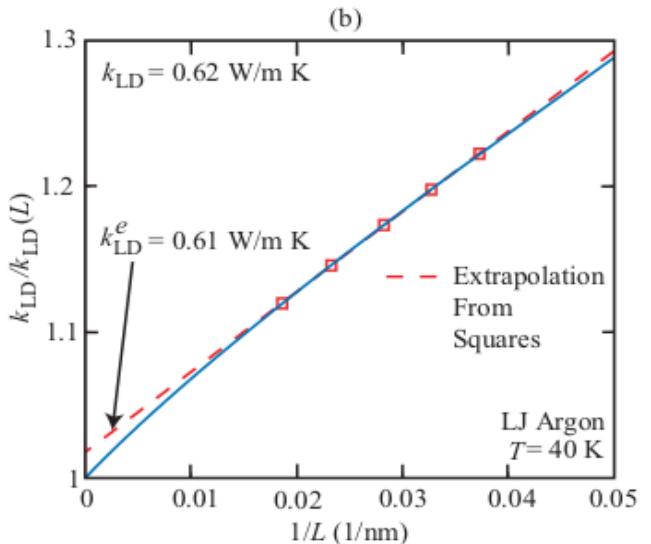
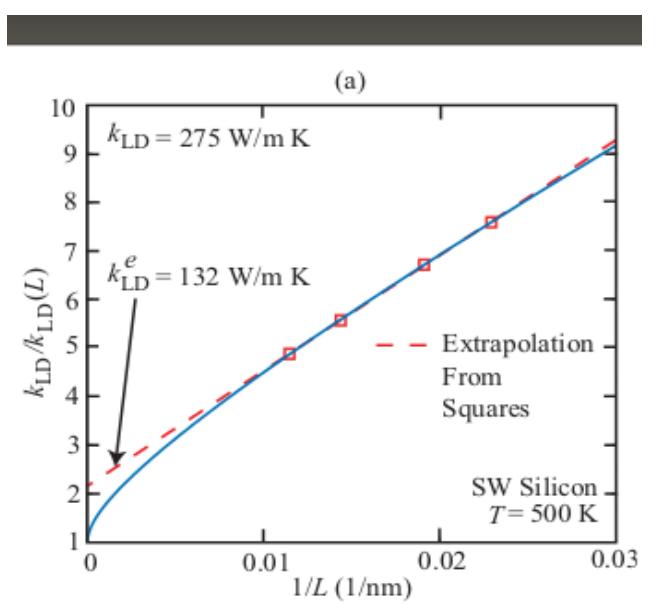
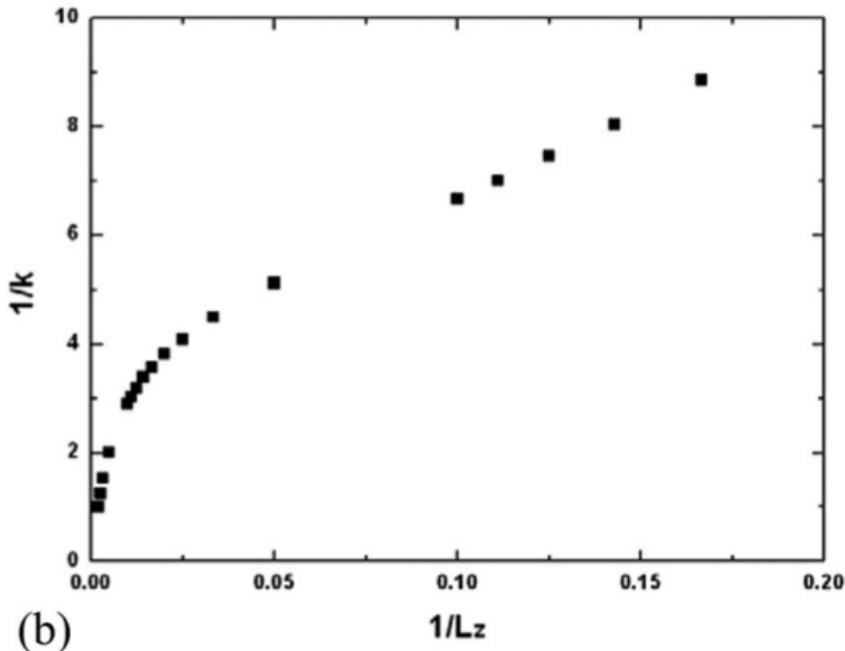
(b)



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# NEMD results

$$\kappa(k_x = k_y = 0) = \frac{C_1 k_B}{3A\pi^2} L_z.$$



PHYSICAL REVIEW B 81, 214305 (2010)

# NEMD results

## Divergent term

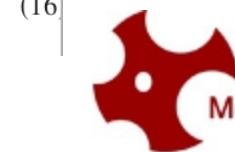
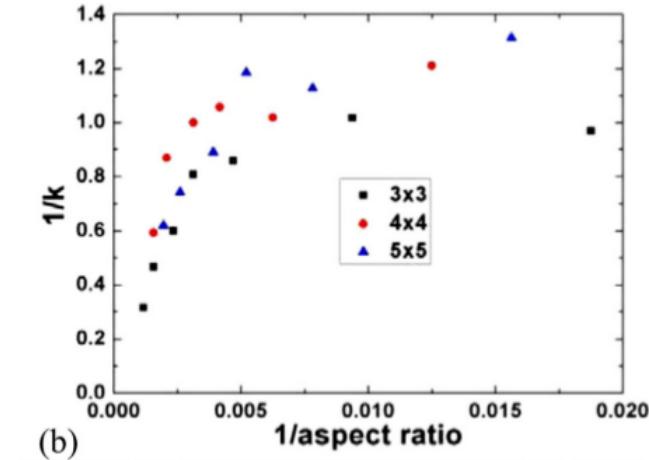
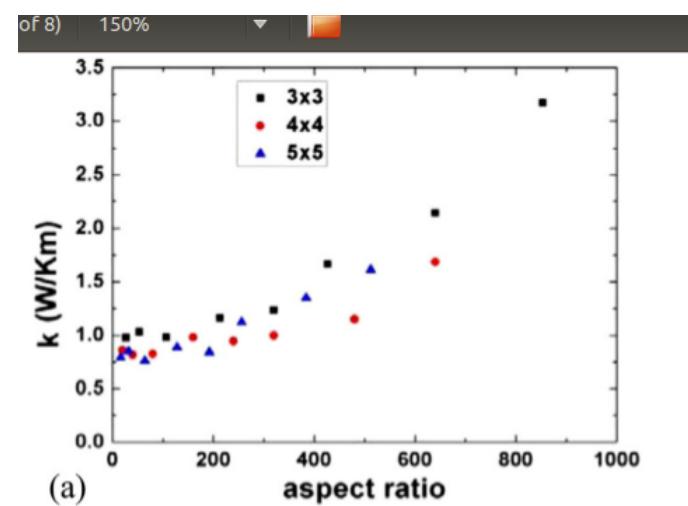
$$\kappa(k_x = k_y = 0) = \frac{C_1 k_B}{3A\pi^2} L_z.$$

## Phonon lifetime from Anharmonic Lattice Dynamics

$$\tau\left(\frac{\boldsymbol{\kappa}}{\nu}\right) = \frac{1}{2\Gamma\left(\frac{\boldsymbol{\kappa}}{\nu}\right)}.$$

$$\begin{aligned} \Gamma\left(\frac{\boldsymbol{\kappa}}{\nu}\right) &= \frac{\pi\hbar}{16N} \sum_{\boldsymbol{\kappa}', \nu'}^{N, 3n} \sum_{\boldsymbol{\kappa}'', \nu''}^{N, 3n} \left| \Phi\left(\frac{\boldsymbol{\kappa}}{\nu}, \frac{\boldsymbol{\kappa}'}{\nu'}, \frac{\boldsymbol{\kappa}''}{\nu''}\right) \right|^2 \left\{ \left[ f\left(\frac{\boldsymbol{\kappa}'}{\nu'}\right) + f\left(\frac{\boldsymbol{\kappa}''}{\nu''}\right) + 1 \right] \left[ \delta\left(\omega\left(\frac{\boldsymbol{\kappa}}{\nu}\right) - \omega\left(\frac{\boldsymbol{\kappa}'}{\nu'}\right) - \omega\left(\frac{\boldsymbol{\kappa}''}{\nu''}\right)\right) \right] + \left[ f\left(\frac{\boldsymbol{\kappa}'}{\nu'}\right) - f\left(\frac{\boldsymbol{\kappa}''}{\nu''}\right) \right] \right. \\ &\quad \times \left. \left[ \delta\left(\omega\left(\frac{\boldsymbol{\kappa}}{\nu}\right) + \omega\left(\frac{\boldsymbol{\kappa}'}{\nu'}\right) - \omega\left(\frac{\boldsymbol{\kappa}''}{\nu''}\right)\right) - \delta\left(\omega\left(\frac{\boldsymbol{\kappa}}{\nu}\right) - \omega\left(\frac{\boldsymbol{\kappa}'}{\nu'}\right) + \omega\left(\frac{\boldsymbol{\kappa}''}{\nu''}\right)\right) \right] \right\}. \end{aligned}$$

PHYSICAL REVIEW B 79, 064301 (2009)



# Finite Simulation effect

PHYSICAL REVIEW B 84, 085204 (2011)

## Phonon lifetime from Anharmonic Lattice Dynamics

$$\tau\left(\begin{array}{c} \boldsymbol{\kappa} \\ \nu \end{array}\right) = \frac{1}{2\Gamma\left(\begin{array}{c} \boldsymbol{\kappa} \\ \nu \end{array}\right)}.$$

There is an additional problem with finite-size MD simulations. Even though momentum is still conserved in a three-phonon process, because the modes are *discrete* in a finite supercell, energy conservation will not always be possible, unless the energy difference  $\omega - \omega_1 - \omega_2 \leq \Gamma$ , where  $\Gamma$  is on the order of the sum of inverse lifetimes of the three considered phonons. If this relation is not satisfied, the considered three-phonon scattering will not take place in a finite supercell, and this will lead to an overestimation of the lifetime of the phonons and, thus, of the thermal conductivity.

These competing effects, namely, an overestimation of  $\kappa$  due to limited phase space for energy conservation and an underestimation due to the cutoff of low-frequency acoustic modes, may lead to a magical cancellation, resulting in thermal conductivities in good agreement with experiments even for moderate supercell size. This error cancellation will likely affect the temperature dependence of  $\kappa$ : at higher temperatures, the discreteness error is reduced as  $\Gamma$  increases linearly with

$$\Gamma\left(\begin{array}{c} \boldsymbol{\kappa} \\ \nu \end{array}\right) = \frac{\pi\hbar}{16N} \sum_{\boldsymbol{\kappa}', \nu'}^{N, 3n} \sum_{\boldsymbol{\kappa}'', \nu''}^{N, 3n} \left| \Phi\left(\begin{array}{ccc} \boldsymbol{\kappa} & \boldsymbol{\kappa}' & \boldsymbol{\kappa}'' \\ \nu & \nu' & \nu'' \end{array}\right) \right|^2 \left\{ \left[ f\left(\begin{array}{c} \boldsymbol{\kappa}' \\ \nu' \end{array}\right) + f\left(\begin{array}{c} \boldsymbol{\kappa}'' \\ \nu'' \end{array}\right) + 1 \right] \left[ \delta\left(\omega\left(\begin{array}{c} \boldsymbol{\kappa} \\ \nu \end{array}\right) - \omega\left(\begin{array}{c} \boldsymbol{\kappa}' \\ \nu' \end{array}\right) - \omega\left(\begin{array}{c} \boldsymbol{\kappa}'' \\ \nu'' \end{array}\right)\right) \right] + \left[ f\left(\begin{array}{c} \boldsymbol{\kappa}' \\ \nu' \end{array}\right) - f\left(\begin{array}{c} \boldsymbol{\kappa}'' \\ \nu'' \end{array}\right) \right] \right. \\ \left. \times \left[ \delta\left(\omega\left(\begin{array}{c} \boldsymbol{\kappa} \\ \nu \end{array}\right) + \omega\left(\begin{array}{c} \boldsymbol{\kappa}' \\ \nu' \end{array}\right) - \omega\left(\begin{array}{c} \boldsymbol{\kappa}'' \\ \nu'' \end{array}\right)\right) - \delta\left(\omega\left(\begin{array}{c} \boldsymbol{\kappa} \\ \nu \end{array}\right) - \omega\left(\begin{array}{c} \boldsymbol{\kappa}' \\ \nu' \end{array}\right) + \omega\left(\begin{array}{c} \boldsymbol{\kappa}'' \\ \nu'' \end{array}\right)\right) \right] \right\}. \quad (16)$$

PHYSICAL REVIEW B 79, 064301 2009

# 3D Anisotropic Crystal

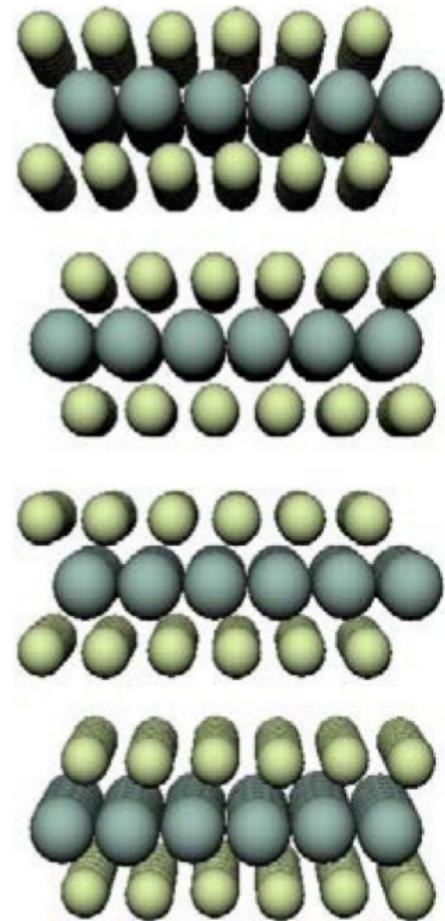
WSe<sub>2</sub>

Anisotropic crystal continuous in z

$$\kappa = D \sum_{k_x} \sum_{k_y} \int_{2\pi/L_z}^{\pi/a} \frac{1}{k_x^2 + k_y^2 + \frac{c_z^2}{c_x^2} k_z^2} dk_z,$$

Divergent term

$$\kappa(k_x = k_y = 0) = D \frac{c_x^2}{c_z^2} L_z.$$

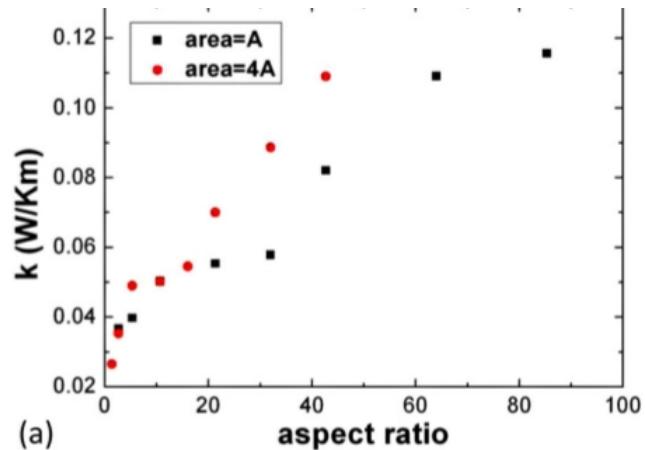


# 3D Anisotropic Crystal

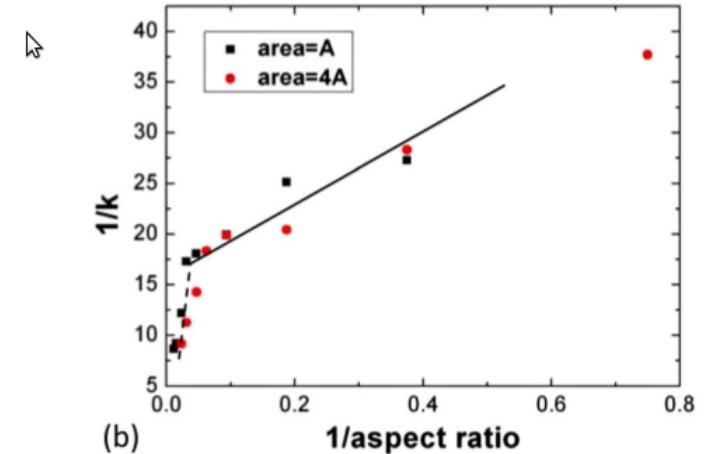
Divergent term

$$\kappa(k_x = k_y = 0) = D \frac{c_x^2}{c_z^2} L_z.$$

WSe<sub>2</sub>



(a) aspect ratio



(b) 1/aspect ratio

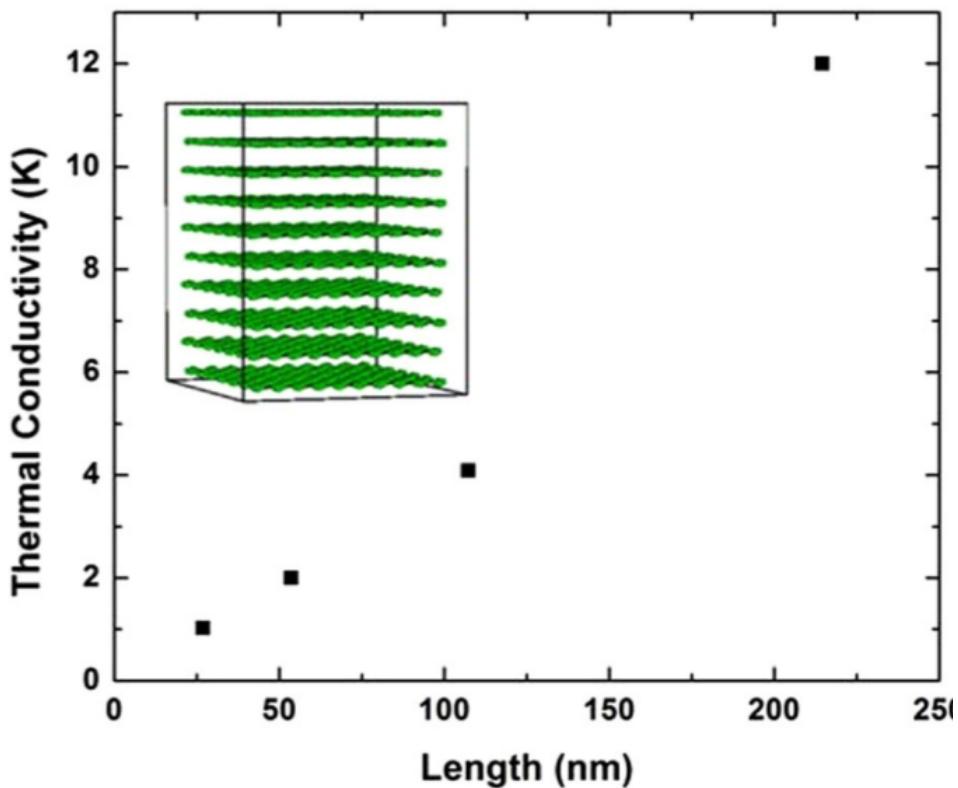
# 3D Anisotropic Crystal

Divergent term

$$\kappa(k_x = k_y = 0) = D \frac{c_x^2}{c_z^2} L_z.$$

Graphite interplane interactions are much weaker than WSe<sub>2</sub>

Graphite

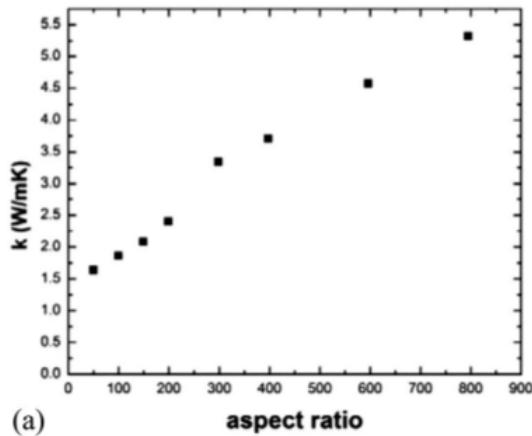


# 1D LJ nanowire

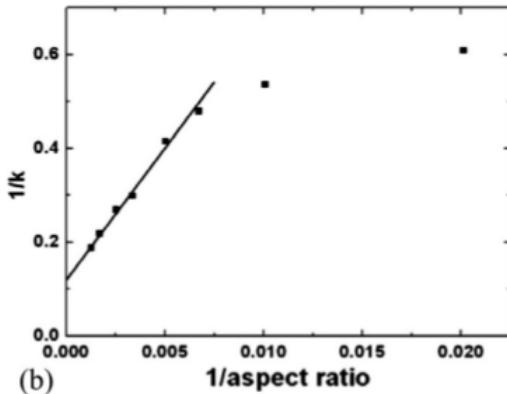
Divergent term

$$\kappa(k_x = k_y = 0) = D \frac{c_x^2}{c_z^2} L_z.$$

1D chains do not have divergent conductivities



(a) aspect ratio



(b)  $1/\text{Aspect Ratio}$

LJ nanowire



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