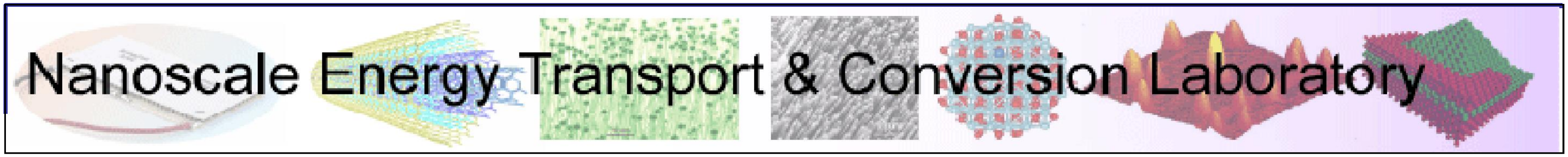

Fundamentals of Phonon Transport Modeling: Formulation, Implementation, and Applications

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ASME IMECE
November 14, 2016

Outline

- Session I (1:30 PM – 3:15 PM)
  1. Introduction (McGaughey)
  2. MD simulation, Green Kubo, direct method (Ruan)
  3. **Harmonic lattice dynamics, spectral methods (Ruan)**

- Session II (3:45 PM – 5:30 PM)
  4. Anharmonic lattice dynamics, first principles (McGaughey)
  5. Phonon-boundary and phonon-defect scattering (McGaughey)
  6. Phonon-electron coupling and non-equilibrium (Ruan)

- **3.1 Harmonic lattice dynamics and phonon dispersion**
- 3.2 Phonon normal mode analysis
- 3.3 Phonon wave packet method

Thermal Radiation: Broad Band Photons

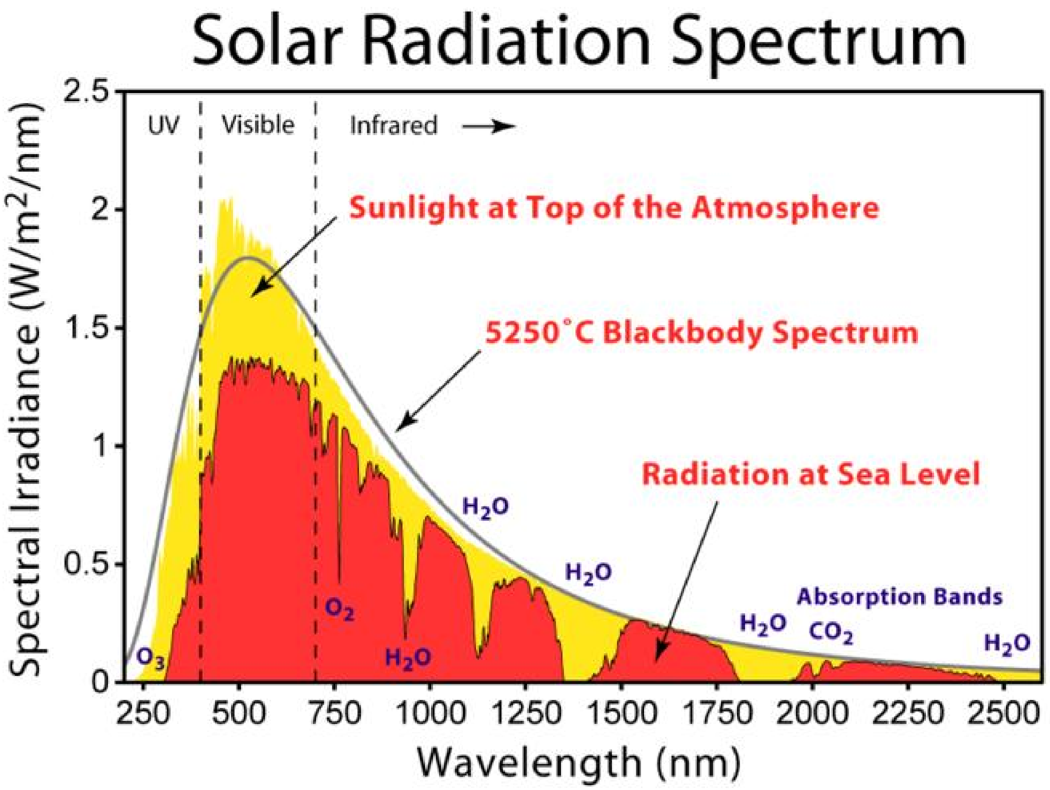


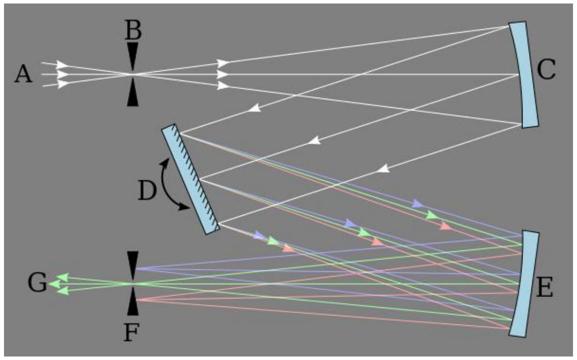
Image of Robert A. Rohde / Global Warming Art

- Planck distribution

$$E_{\lambda,b}\left(\lambda,T\right)=\frac{C_1}{\lambda^5\left[\exp\left(C_2/\lambda T\right)-1\right]}$$

- Stefan-Boltzmann Law

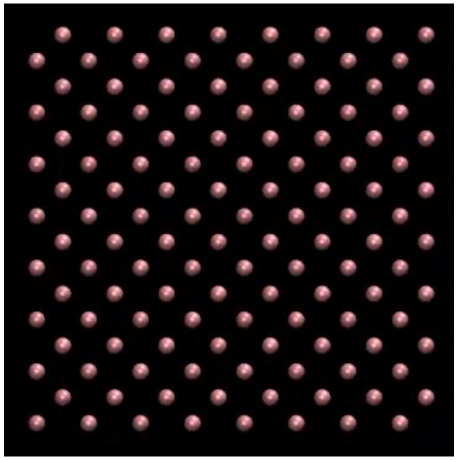
$$E_b=\int_0^\infty E_{\lambda,b}\left(\lambda,T\right)d\lambda=\sigma T^4$$

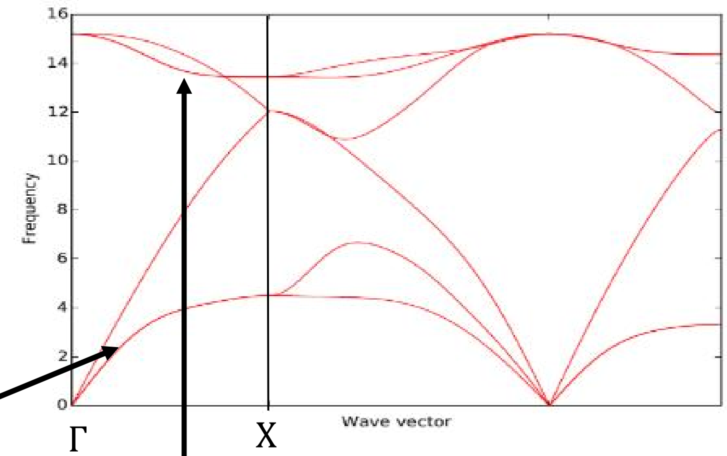


Monochromator

Heat Conduction: Broad Band Phonons

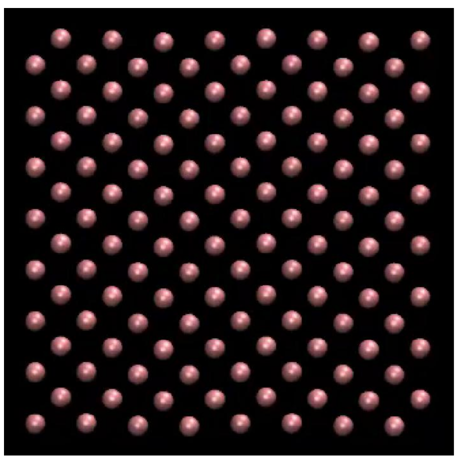
MD Trajectory



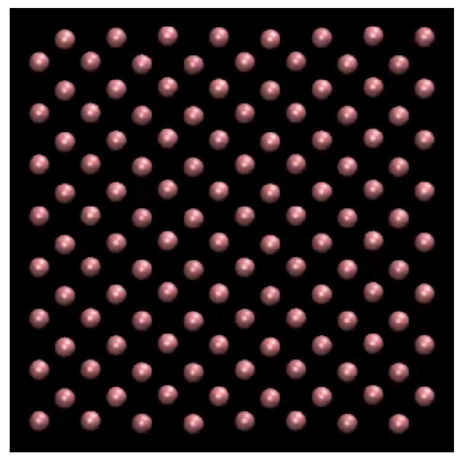


Acoustic Mode

Optical Mode



Mode 1

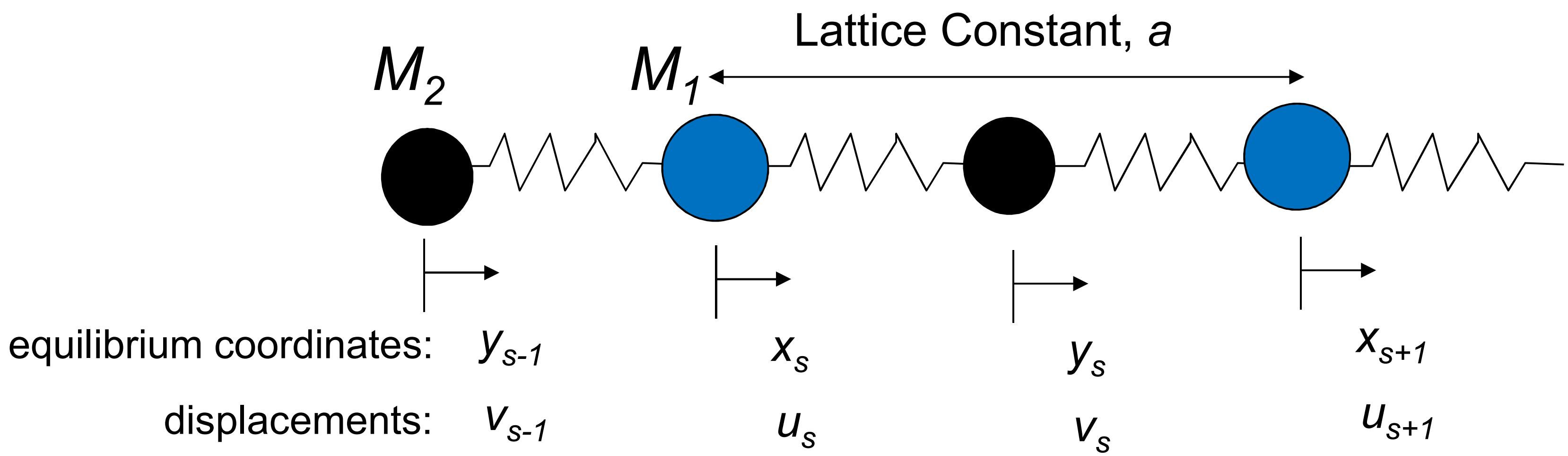


Mode 2

. . .

$$k_x = \frac{1}{V}\sum_{\lambda} c_{\lambda} v_{\lambda,x}^2 \tau_{\lambda}$$

Two-Atom Chain



- Equation of motion:

$$M_1 \frac{d^2 u_s}{dt^2} = C(v_s + v_{s-1} - 2u_s); \qquad (1)$$

$$M_2 \frac{d^2 v_s}{dt^2} = C(u_{s+1} + u_s - 2v_s); \qquad (2)$$

Solution of the Wave Equation

- Assume a wave solution in the form

$$u_s = u_0 \exp(isKa)\exp(-i\omega t)$$
$$v_s = v_0 \exp(isKa)\exp(-i\omega t)$$

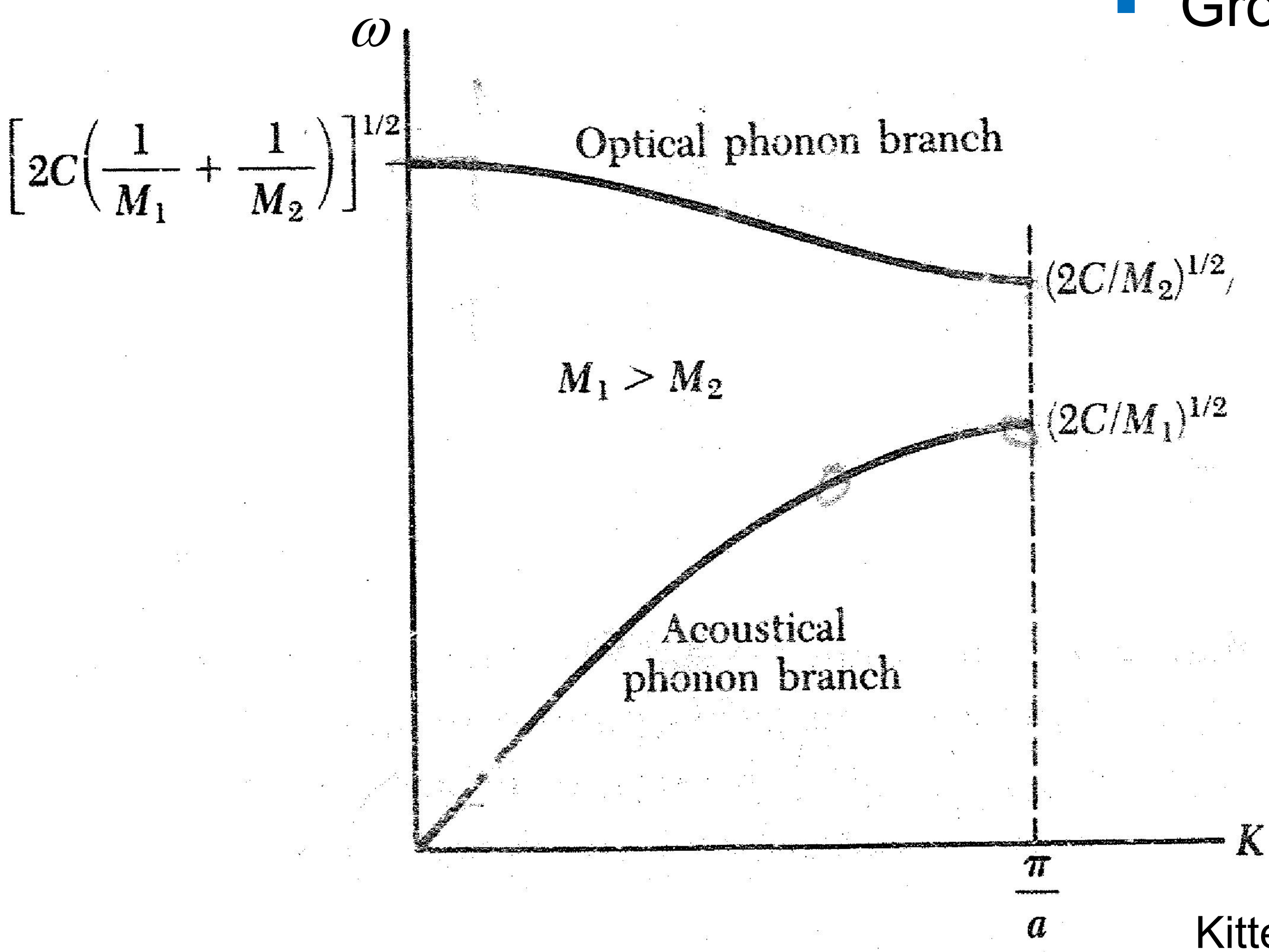
- Then the equations of motion becomes

$$-\omega^2 M_1 u_0 = Cv_0\left[1+\exp(-iKa)\right] - 2Cu_0$$
$$-\omega^2 M_2 v_0 = Cu_0\left[\exp(iKa)+1\right] - 2Cv_0$$

- The homogeneous linear equations have a solution only if

$$\begin{vmatrix} 2C - M_1\omega^2 & -C\left[1+\exp(-iKa)\right] \\ -C\left[1+\exp(iKa)\right] & 2C - M_2\omega^2 \end{vmatrix} = 0$$

Dispersion Relation



- Group Velocity:

$$v_g = \frac{d\omega}{dK}$$

Kittel, Introduction to Solid State Physics.

Dynamical Matrix Approach

$$\det\begin{bmatrix} 2C - M_1\omega^2 & -C\left[1+\exp(-iKa)\right] \\ -C\left[1+\exp(iKa)\right] & 2C - M_2\omega^2 \end{bmatrix} = 0$$

or

$$\det\left\{\begin{bmatrix} \frac{2C}{M_1} & \frac{-C}{\sqrt{M_1M_2}}\left[1+\exp\left(-iKa\right)\right] \\ \frac{-C}{\sqrt{M_1M_2}}\left[1+\exp\left(iKa\right)\right] & \frac{2C}{M_2} \end{bmatrix} - \omega^2\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\right\} = 0$$

- Define

$$D(K) = \begin{bmatrix} \frac{2C}{M_1} & \frac{-C}{\sqrt{M_1M_2}}\left[1+\exp\left(-iKa\right)\right] \\ \frac{-C}{\sqrt{M_1M_2}}\left[1+\exp\left(iKa\right)\right] & \frac{2C}{M_2} \end{bmatrix}$$

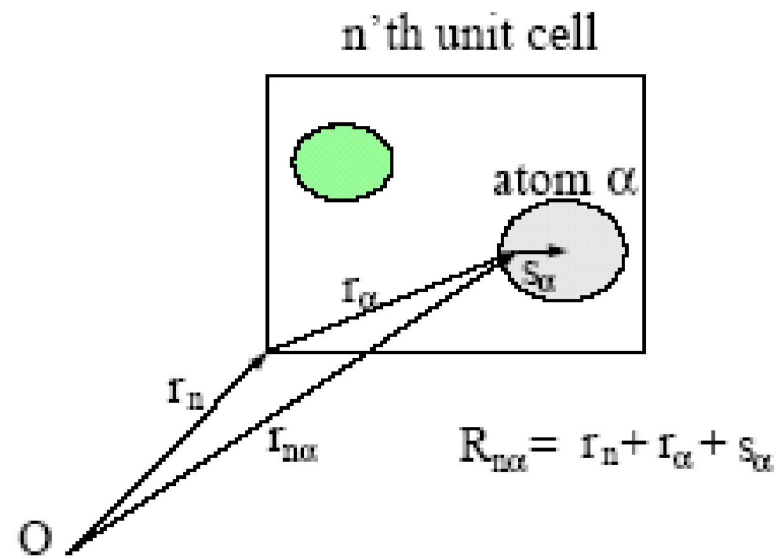
Dynamical Matrix

- So

$$\det\left[D(K) - \omega^2 I\right] = 0$$

Atomic Position Nomenclature in 3D

- An atom in unit cell *n* and at basis position α has an equilibrium position $r_{n\alpha}$
- Subscripts *i* and *j* denote direction
- Subscripts *m* and β denote the position of a different atom in the lattice (analogous to *n* and α)
- Perturbed expression for the potential ϕ (like a Taylor series expansion)



$$\Phi_{n\alpha i}^{m\beta j} = \frac{\partial^2 \phi}{\partial r_{n\alpha i} \partial r_{m\beta j}}$$

$$\phi(\{r_{n\alpha i} + s_{n\alpha i}\}) = \phi(\{r_{n\alpha i}\}) + \frac{\partial \phi}{\partial r_{n\alpha i}} s_{n\alpha i} + \frac{1}{2} \frac{\partial^2 \phi}{\partial r_{n\alpha i} \partial r_{m\beta j}} s_{n\alpha i} s_{m\beta j}$$

0 because assumed zero potential and first derivative at the equilibrium position

Equations of Motion

- The force on the atom at position $n\alpha$ along direction i can be expressed in terms of the spatial derivative of potential energy

$$F_{n\alpha i} = -\frac{\partial \phi(\mathbf{r}_{n\alpha i} + \mathbf{s}_{n\alpha i})}{\partial \mathbf{s}_{n\alpha i}} = -\Phi_{n\alpha i}^{m\beta j}\mathbf{s}_{m\beta j} = M_\alpha \ddot{\mathbf{s}}_{n\alpha i}$$

- Atomic displacement s can be expressed as a Fourier wave

$$\mathbf{s}_{n\alpha i} = \frac{1}{\sqrt{M_\alpha}} u_{\alpha i}(\mathbf{K}) \exp\left(-i\omega t + i\mathbf{K}\cdot\mathbf{r}_n\right)$$

  - The wave vector **K** is an important parameter (inversely proportional to wavelength)
  - Wave frequency ω gives the rate of vibration of the wave

- Substituting into the equation of motion, we find

$$\omega^2 u_{\alpha i}(\mathbf{K}) = \frac{1}{\sqrt{M_\alpha M_\beta}} \Phi_{n\alpha i}^{m\beta j} \exp\left[i\mathbf{K}\cdot(\mathbf{r}_m - \mathbf{r}_n)\right] u_{\beta j}(\mathbf{K})$$

Dynamical Matrix and Dispersion Relation

- The dynamical matrix contains each “spring constant”

$$D_{\alpha i}^{\beta j} = \frac{1}{\sqrt{M_\alpha M_\beta}} \Phi_{n\alpha i}^{m\beta j} \exp\left[i\mathbf{K}\cdot(\mathbf{r}_m - \mathbf{r}_n)\right] = \frac{1}{\sqrt{M_\alpha M_\beta}} \Phi_{0\alpha i}^{p\beta j} \exp\left[i\mathbf{K}\cdot\mathbf{r}_p\right]$$

  - Second equality from translational invariance, and $\mathbf{r}_p$ is defined as $\mathbf{r}_m - \mathbf{r}_n$
- The euqation of motion becomes

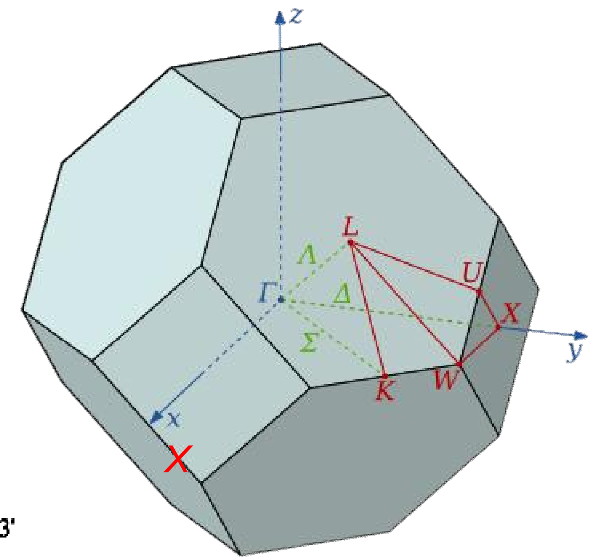
$$\left(D_{\alpha i}^{\beta j} - \omega^2 \delta_{\alpha i}^{\beta j}\right) u_{\beta j}(\mathbf{K}) = 0$$

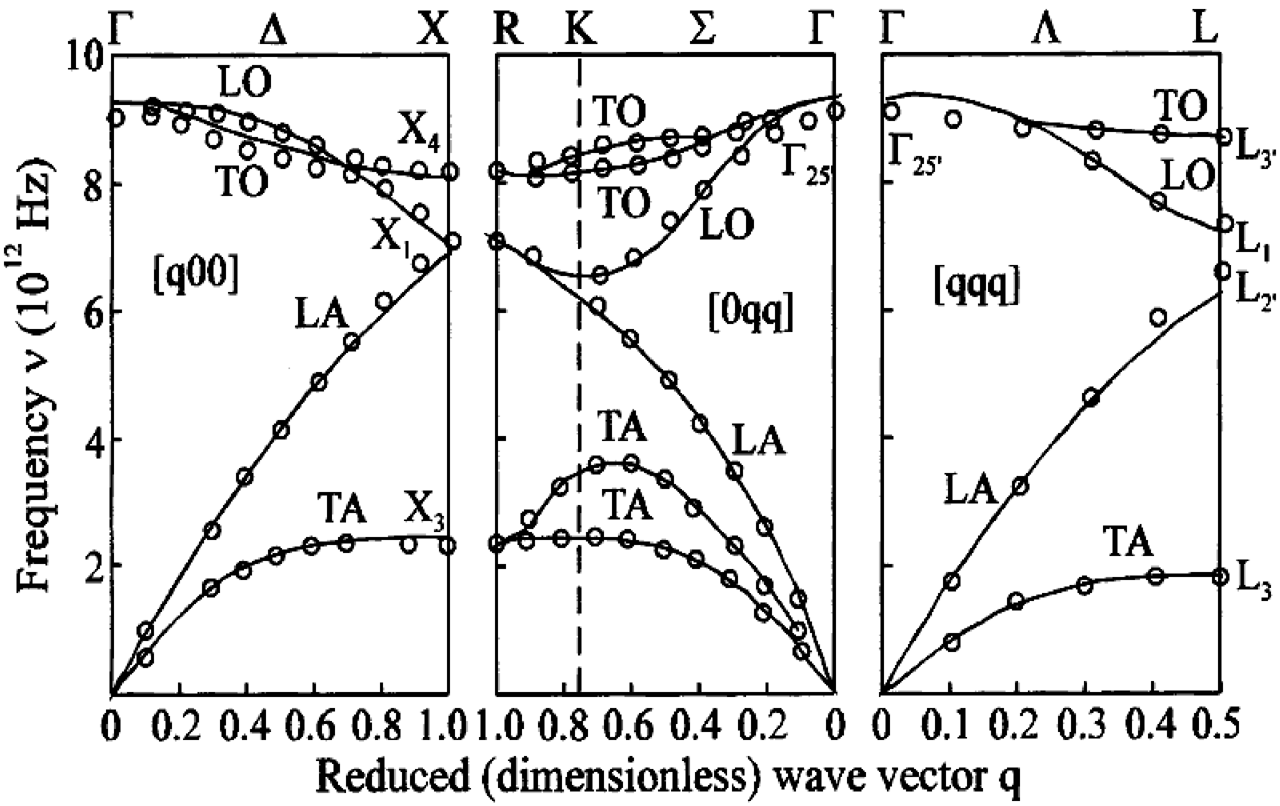
- Where δ is the Kronecker delta (identity matrix).
- The dispersion relation defines the relationship between the wave vector **K** and frequency ω and emerges from the secular equation,

$$\det[\mathbf{D}(\mathbf{K}) - \omega^2\mathbf{I}] = 0$$

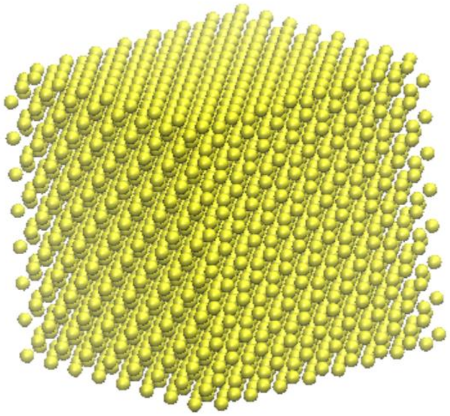
Phonon Dispersion of Silicon

- Phonon dispersion is typically plot along high symmetry lines in the first Brillouin zone.





Spectral Phonon Properties



N atoms

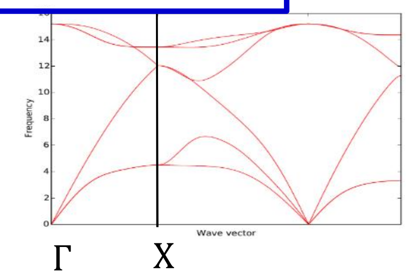
3N vibrational modes

ν: dispersion branch

$\mathbf{k}$: wave vector

Group velocity

$$\boldsymbol{v} = \frac{d\omega}{d\mathbf{k}}, \; v_x = \boldsymbol{v} \cdot \hat{\boldsymbol{x}}$$



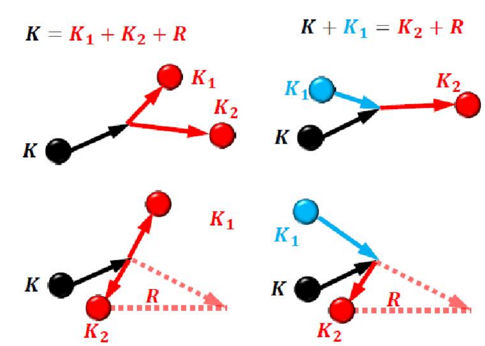
$$\kappa_x = \sum_{\nu,\mathbf{k}} c_{\boxed{\nu,\mathbf{k}}} v_{\nu,\mathbf{k},x}^2 \tau_{\nu,\mathbf{k}}$$

Specific heat

$$c_{\nu,\mathbf{k}} = \hbar\omega \frac{\partial n^0}{\partial T}$$

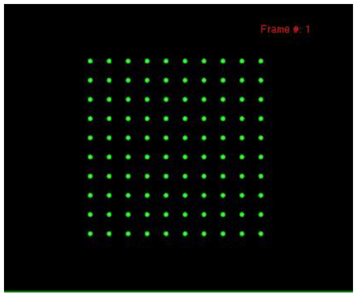
$$n^0 = \frac{1}{\exp(\frac{\hbar\omega}{k_B T}) - 1}$$

Relaxation time $\tau_{\nu,\mathbf{k}}$



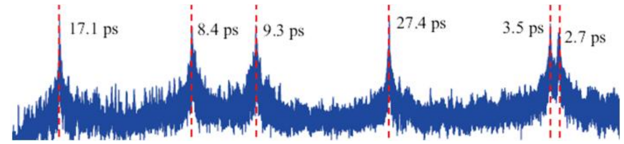
- 3.1 Harmonic lattice dynamics and phonon dispersion
- **3.2 Phonon normal mode analysis**
- 3.3 Phonon wave packet method

Phonon Normal Mode Analysis



MD simulations

Atomic velocity



ω τ

broader peak → shorter τ

Spectral energy density function

$$\Phi(\mathbf{k},\omega)=\sum_{\nu}^{3n}\left|\sum_{\alpha}^{3}\sum_{b}^{n}\int_{0}^{\infty}\sum_{l}^{N_c}\sqrt{\frac{m_b}{N_c}}\boxed{\dot{u}_{\alpha}^{l,b}(t)}\boxed{e_{\alpha}^{*b}(\mathbf{k},\nu)}\exp(-i[\omega t-\mathbf{k}\cdot\mathbf{r}])dt\right|^2$$

Molecular dynamics or lattice dynamics → Eigenvector

Second version without eigenvector:

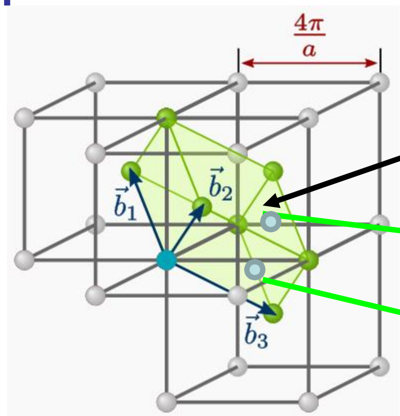
$$\Phi'(\mathbf{k},\omega)=\sum_{\alpha}^{3}\sum_{b}^{n}\left|\sum_{l}^{N_c}\int_{0}^{\infty}\sqrt{\frac{m_b}{N_c}}\dot{u}_{\alpha}^{l,b}(t)\exp(-i[\omega t-\mathbf{k}\cdot\mathbf{r}])dt\right|^2$$

We proved that $\Phi \equiv \Phi'$. Feng, Qiu, Ruan, *JAP*, 117 (19), 195102 (2015)

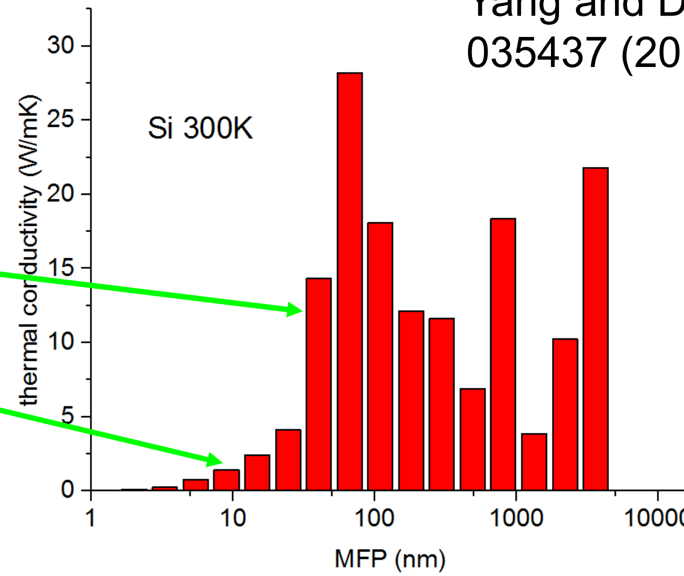
References:

1. A. J. C. Ladd, B. Moran, and W. G. Hoover, Phys. Rev. B 34, 5058 (1986). 2. A.J.H. McGaughey and M. Kaviany, Phys. Rev. B (2004). 3. A. S. Henry and G. Chen, J. Comput. Theor. Nanosci. 5,1 (2008). 4. N. de. Koker, Phys. Rev. Lett. 103, 125902 (2009). 5. Thomas, Turney, Iutzi, Amon, and McGaughey, Phys. Rev. B 81, 081411 (2010). 6. Qiu and Ruan, Appl. Phys. Lett. 100, 193101 (2012). 7. Feng and Ruan, J. Appl. Phys. **117**, 195102 (2015). 8. Feng, Ruan, Ye, Cao, Phys. Rev. B 91, 224301 (2015).

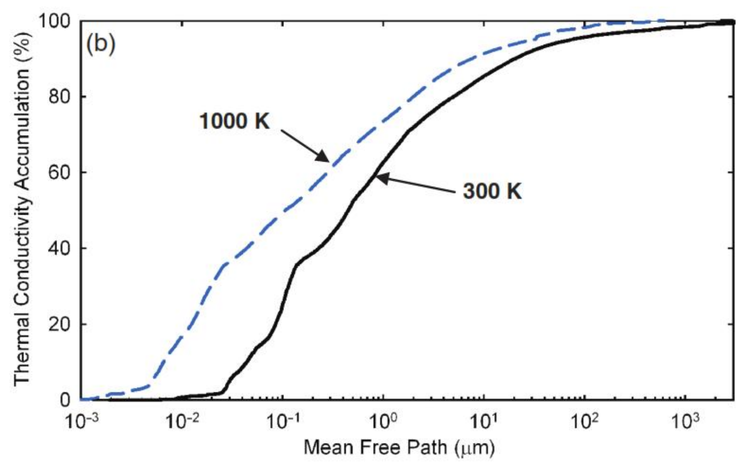
Spectral and Accumulated k v.s. MFP



Yang and Dames, *PRB* **87**, 035437 (2013).

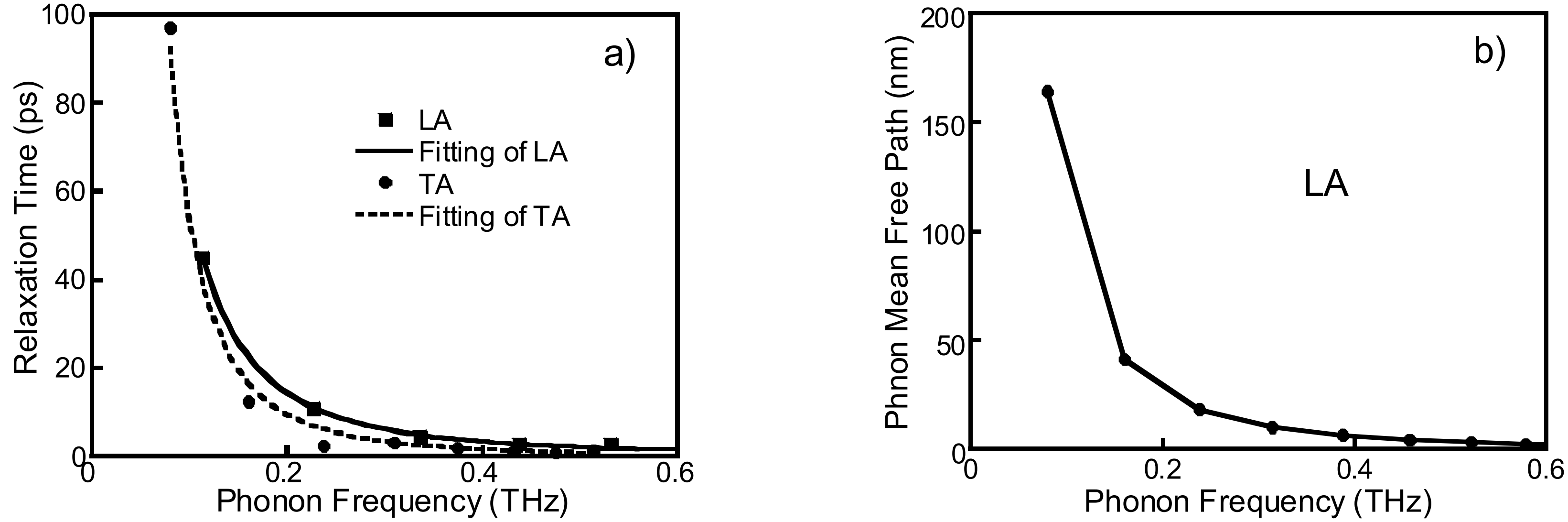


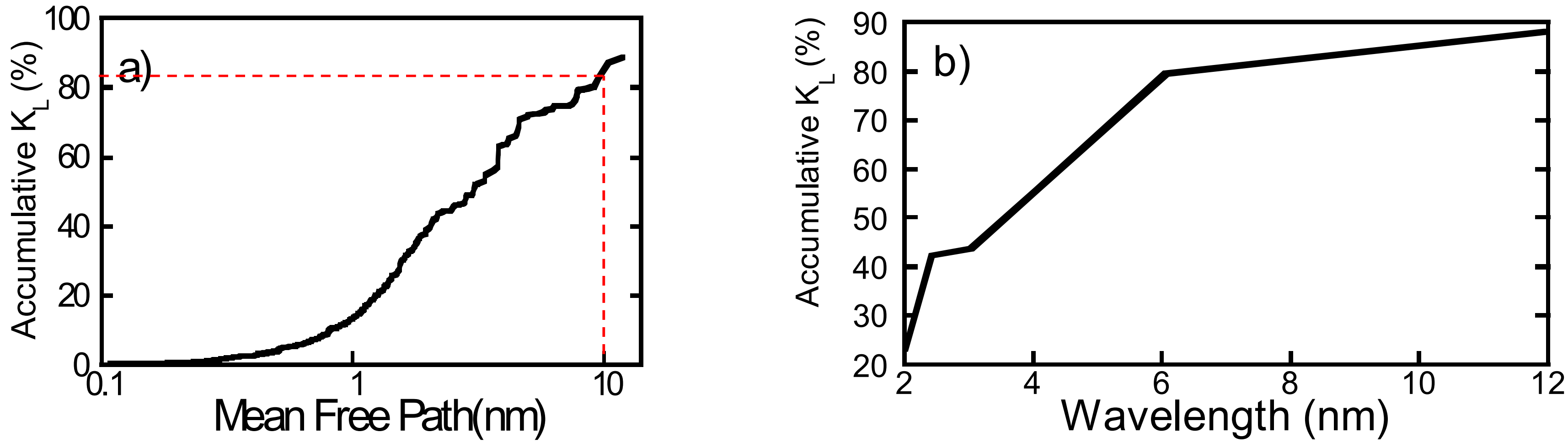
- Discretize MFP into bins
- For each phonon mode, find its MFP and assign it to a bin, add its spectral k to the spectral k of that bin.
- Accumulate the spectral k over MFP.



Henry and Chen, *JCTN* **5**,1-12, (2008).

Relaxation Time and Mean Free Path for Bi_2Te_3

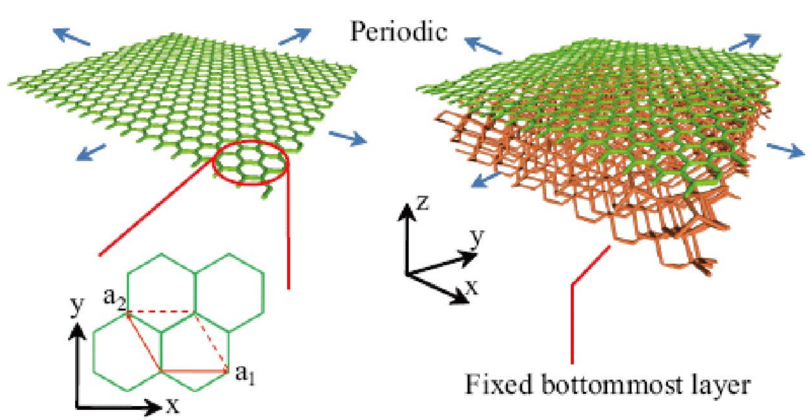




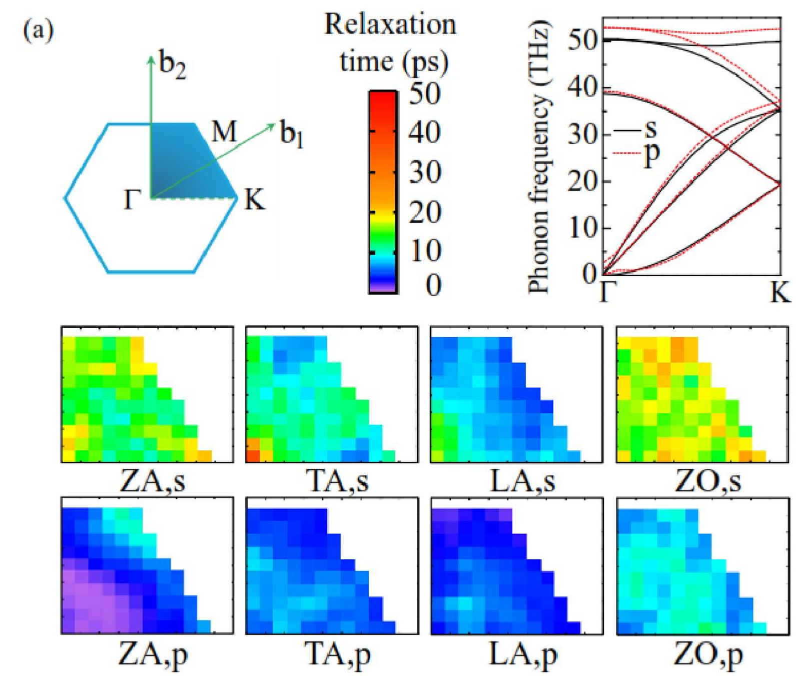
Wang, Qiu, McGaughey, Ruan, and Xu, *J. Heat Transfer* 135, 091102 (2013).

Total phonon scattering rate for perturbed systems

- Small perturbation to bulk system: substrate, impurity, etc.
- Assumption: the perturbation little affects the phonon dispersion, but affects the phonon scattering rates.
- The total scattering rate is calculated for multiple phonon scattering mechanisms without touching the detailed interplay process.



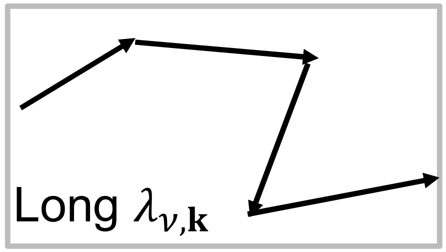
Qiu and Ruan, Appl. Phys. Lett. 100, 193101 (2012).

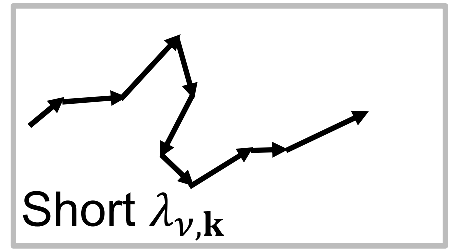


Spectral and Gray Approaches

- Mean free path

$$\lambda_{\nu,\mathbf{k}} = v_{\nu,\mathbf{k}}\tau_{\nu,\mathbf{k}}$$





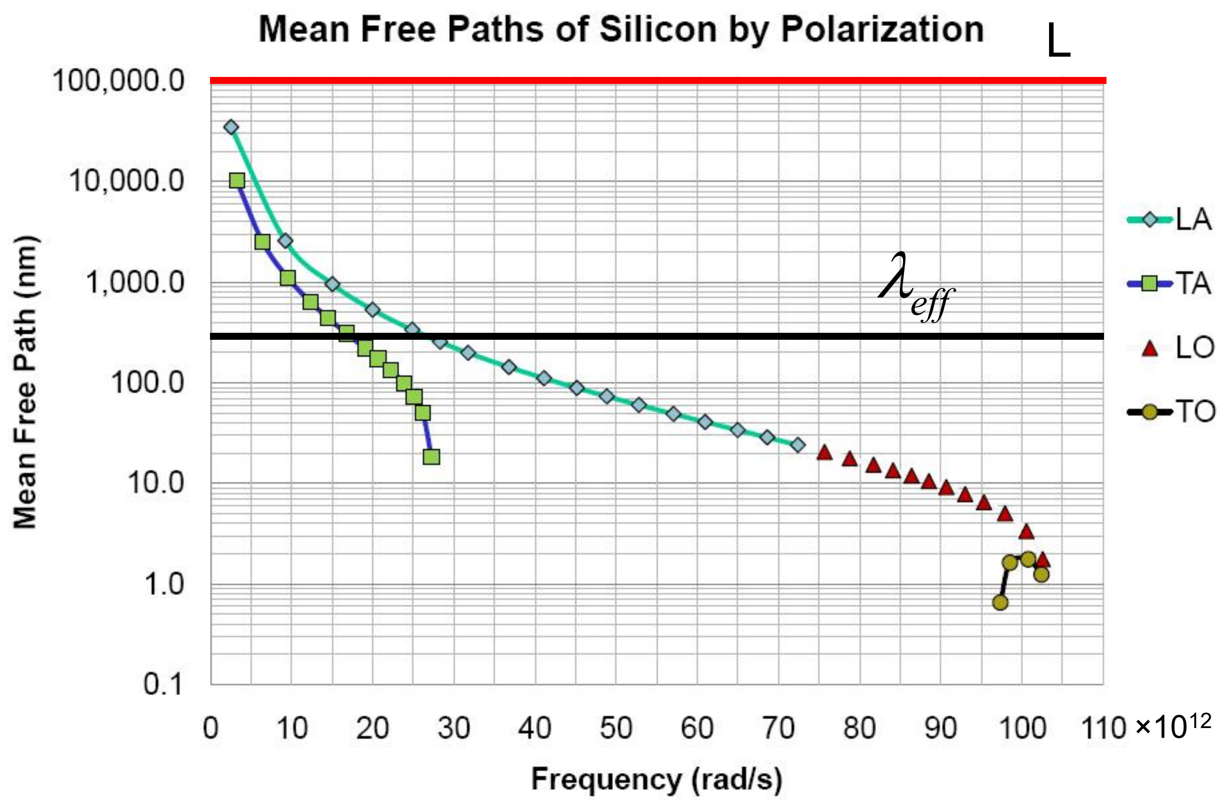
- Spectral approach

$$\kappa_x = \sum_{\nu,\mathbf{k}} c_{\nu,\mathbf{k}} v_{\nu,\mathbf{k},x}^2 \tau_{\nu,\mathbf{k}}$$

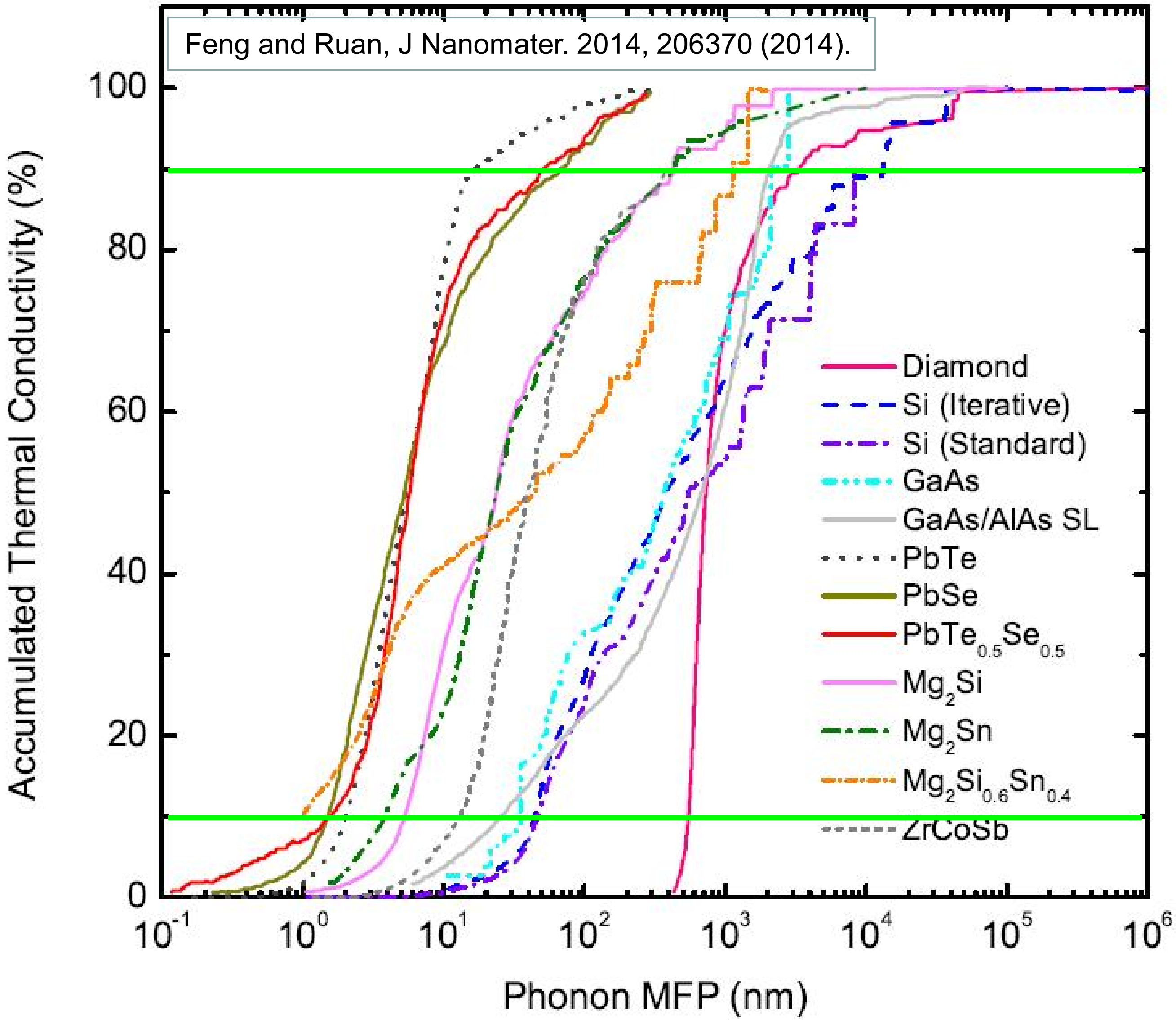
- Gray approach

$$\kappa_x = \frac{1}{3} c v \lambda_{eff}$$

$$= \frac{1}{3} c v^2 \tau_{eff}$$



Thermal conductivity accumulation

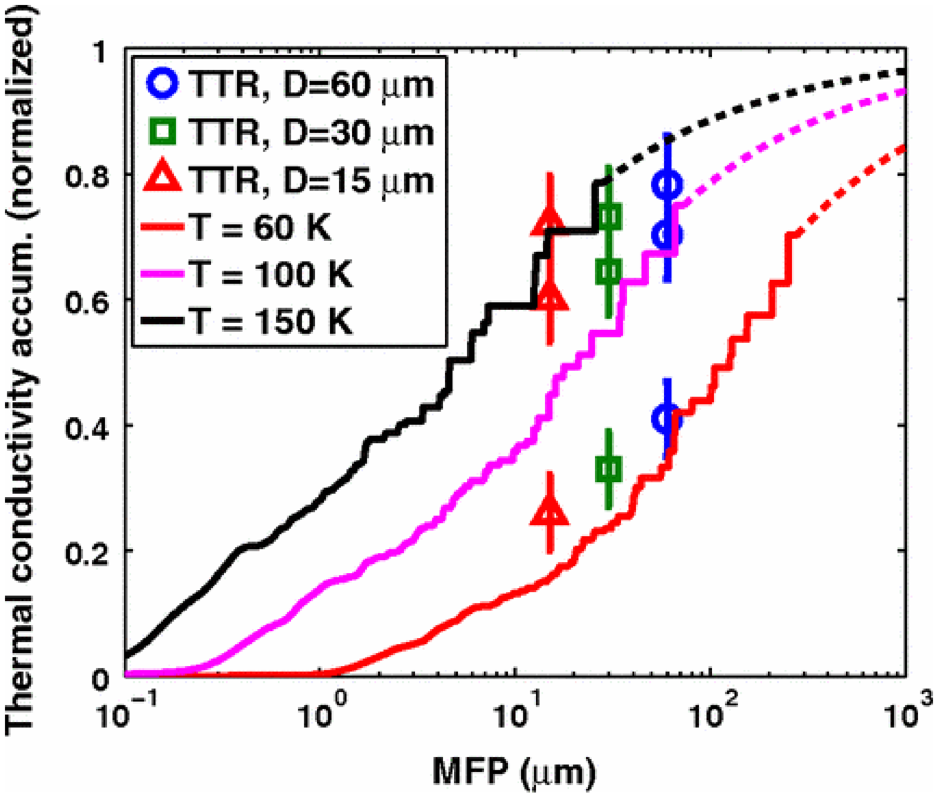


Phonon mean free path in nanostructures

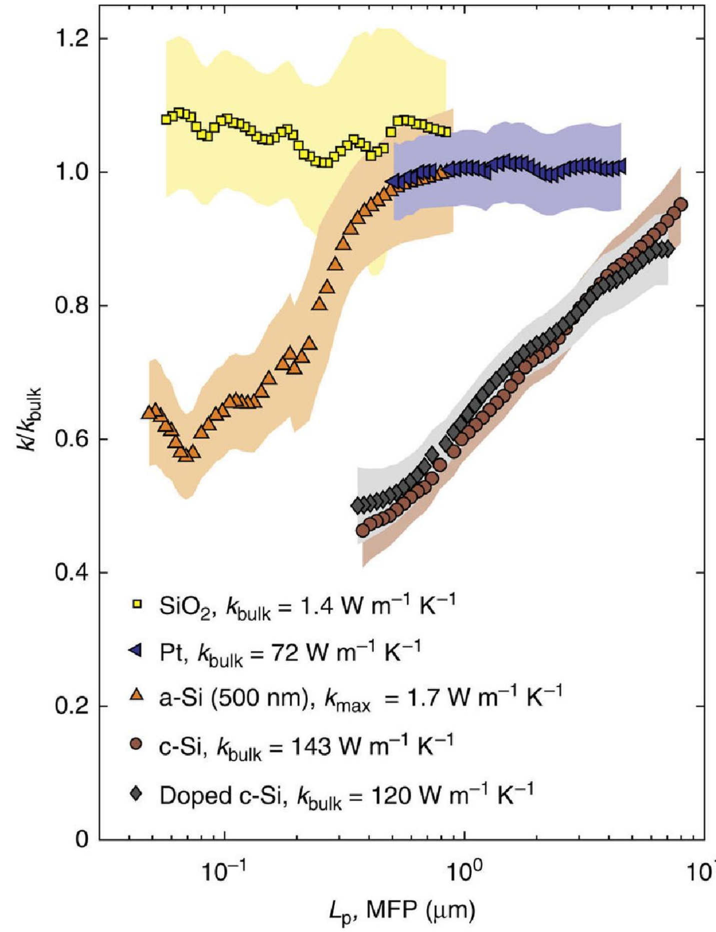
$$\frac{1}{\lambda_p} = \frac{1}{\lambda_{\nu,\mathbf{k}}} + \frac{1}{L}$$

L needs to be comparable to or smaller than $\lambda_{\nu,\mathbf{k}}$ for size effect.

Thermal conductivity accumulation: experiment



Minnich, Johnson, Schmidt, Esfarjani, Dresselhaus, Nelson, and Chen, Phys. Rev. Lett. 107, 095901 (2011)



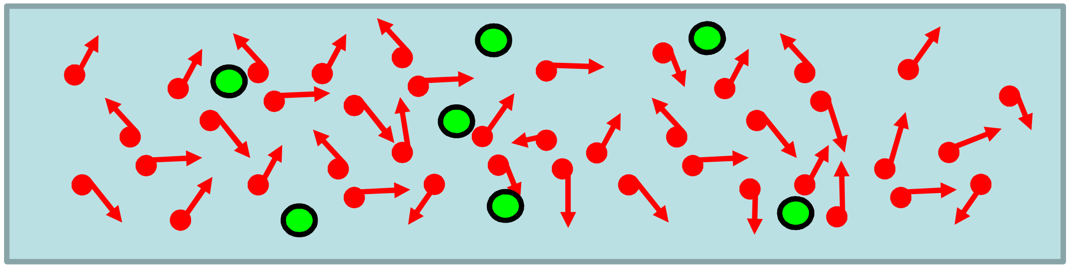
Regner, Sellan, Su, Amon, McGaughey, Malen, Nature communications 4, 1640 (2013)

Other Modal Analysis Techniques

- Modal Heat Flux Decomposition in NEMD:
  - Zhou, Zhang, Hu, PRB 92, 195204 (2015).
- Modal decomposition based on Green-Kubo
  - Lv, Henry, New J. Phys. 18, 013028 (2016).

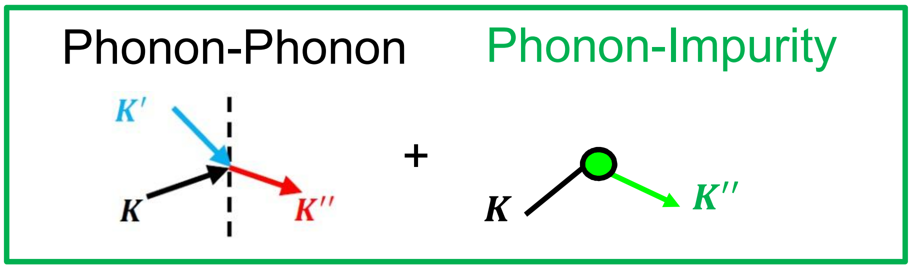
Spectral Matthiessen's Rule revisited

Non-metal



Heat carriers: phonons

Impurities exist everywhere: isotopes, Si_xGe_{1-x}, $PbTe_xSe_{1-x}$, $Bi_2Te_{3-x}Se_x$, etc.

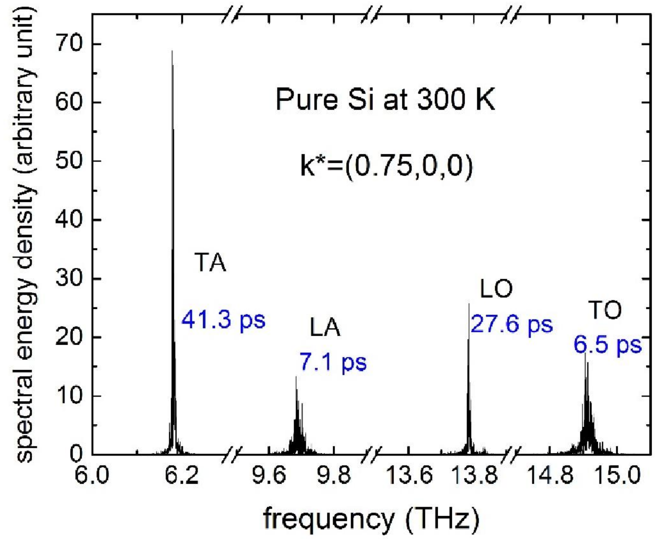


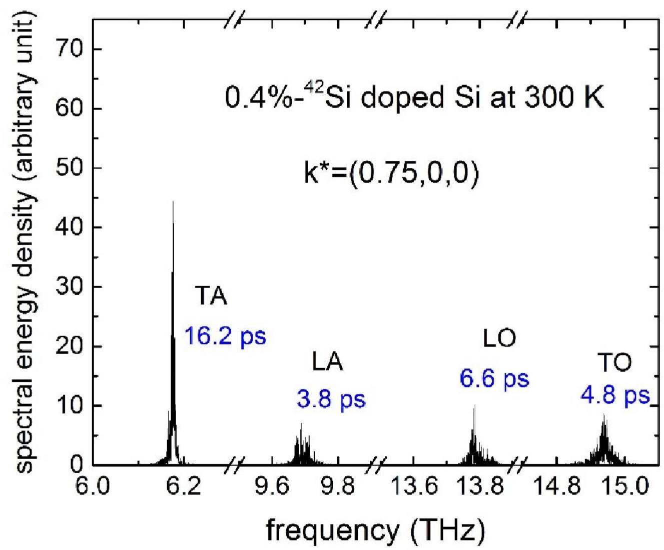
- Spectral Matthiessen's Rule:

$$\tau_{\lambda,tot}^{-1} \overset{?}{=} \tau_{\lambda,p-p}^{-1} + \tau_{\lambda,p-i}^{-1}$$

λ: phonon mode

Total phonon scattering rate without and with impurity doping

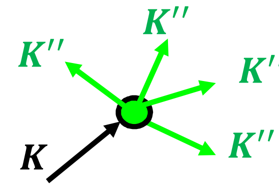




Phonon-impurity scattering rate

$$\tau_{\lambda,i}^{-1} = \frac{\pi}{2N_c}\omega_\lambda^2 \sum_b^n \sum_{\lambda' \neq \lambda} g_b \left| \boldsymbol{e}_b^{\lambda} \cdot \boldsymbol{e}_b^{\lambda' *} \right|^2 \delta(\omega_\lambda - \omega_{\lambda'})$$

$$= \frac{\pi g}{2}\omega_\lambda^2 D(\omega)$$

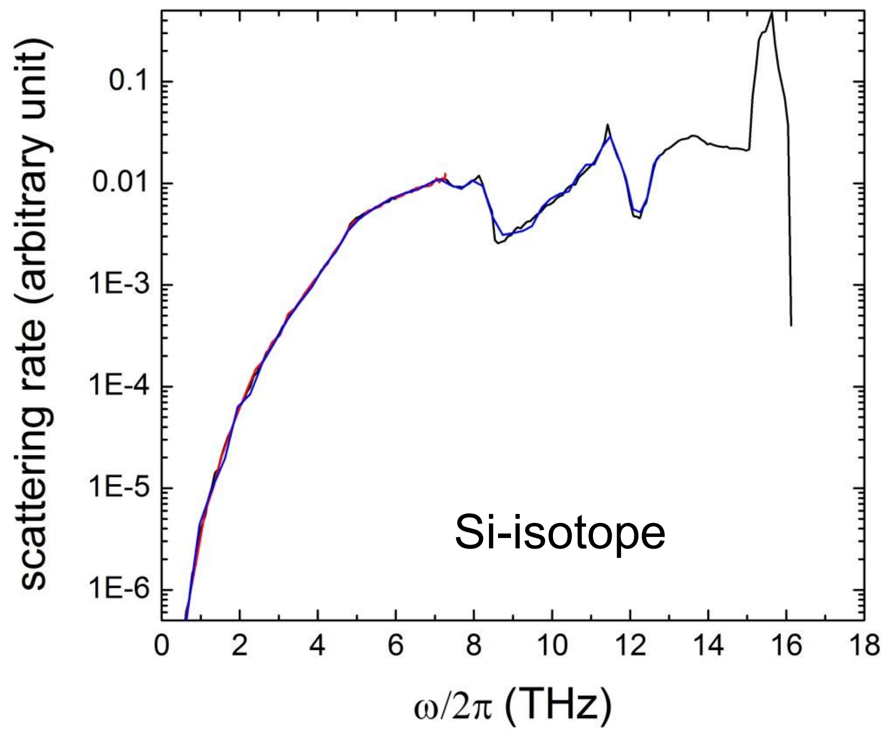


$$g = \sum_i n_i \left(\frac{\Delta M_i}{M}\right)^2$$ Mass perturbation

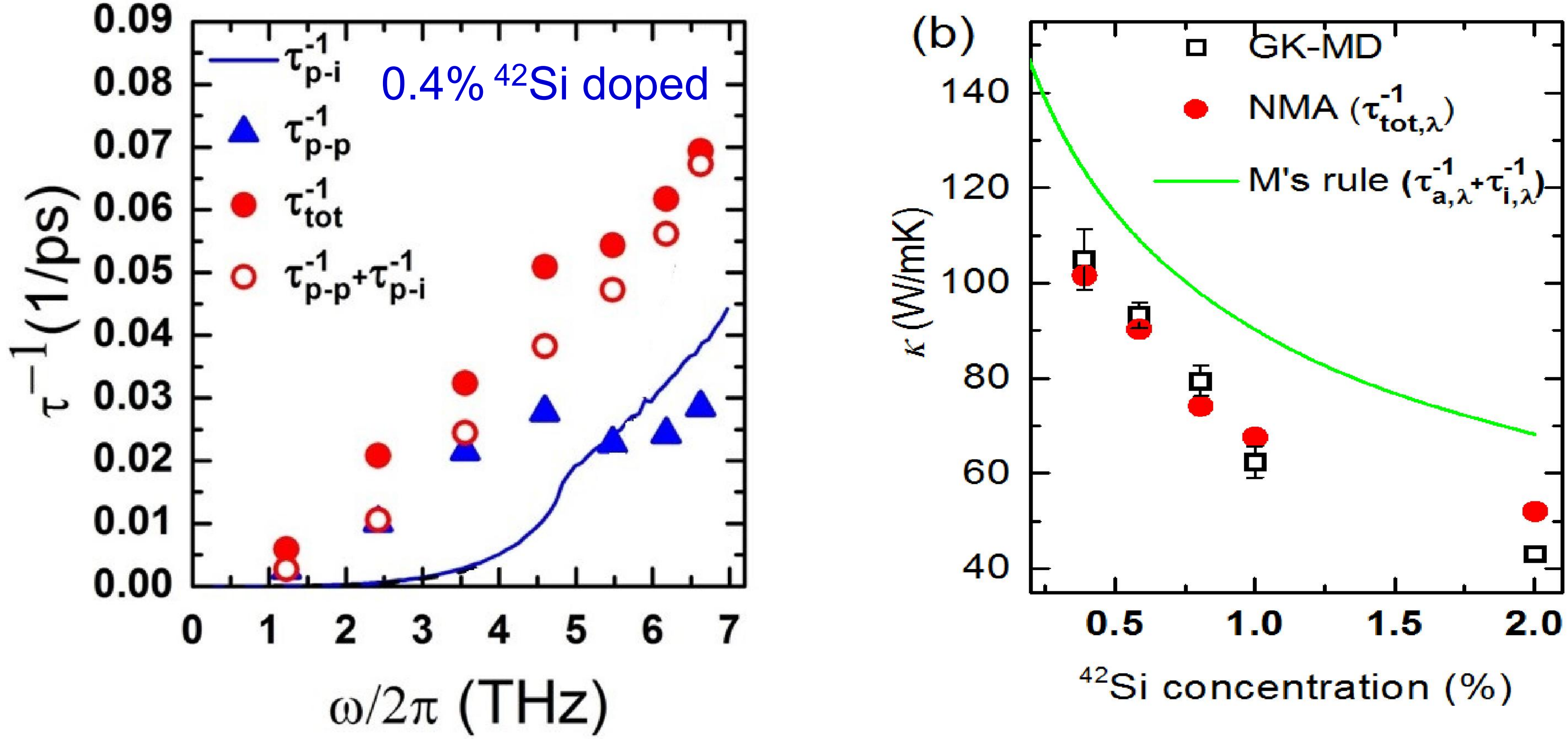
$D(\omega)$: Phonon density of states

S. Tamura, Phys. Rev. B 27, 858 (1983)

Impurity scattering rate



Scattering rates and thermal conductivity

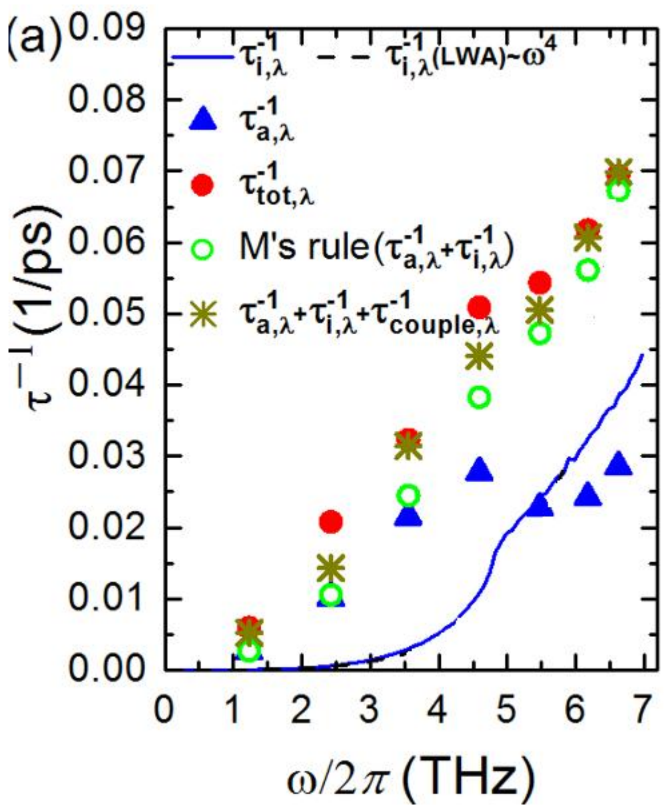


Feng, Qiu, and Ruan, Phys. Rev. B. 92, 235206 (2015).

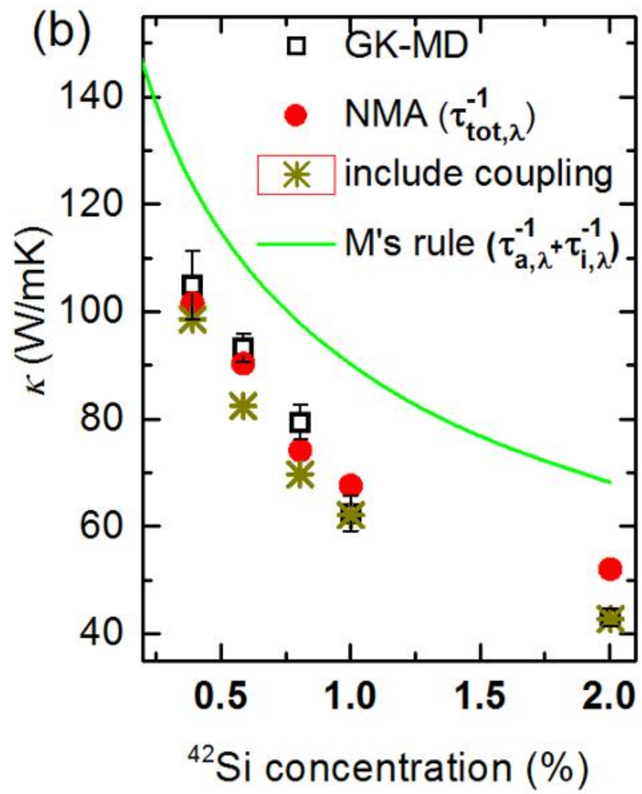
Coupling in Isotope-Doped Silicon

Bulk Silicon 300K

0.4% ^{42}Si doped



0.4% - 2% ^{42}Si doped



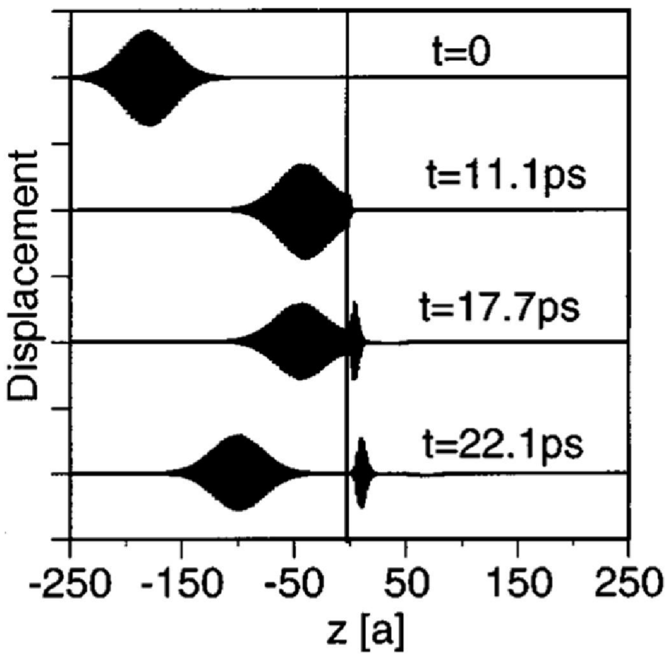
- $\tau_a^{-1} + \tau_i^{-1} + \tau_{couple}^{-1} \sim \tau_{tot}^{-1}$

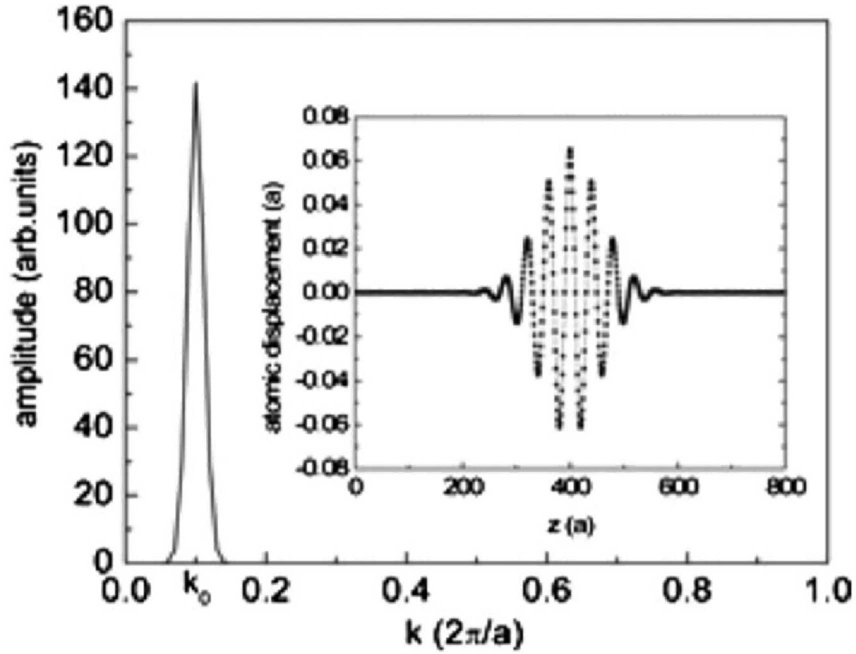
Feng, Qiu, and Ruan, Phys. Rev. B. 92, 235206 (2015).

- k from $\tau_a^{-1} + \tau_i^{-1} + \tau_{couple}^{-1}$ agrees well with GK and NMA

- Harmonic lattice dynamics and phonon dispersion
- Phonon normal mode analysis
- **Phonon wave packet method**

Phonon Wave-packet Method





Displacement of the ith atom in the lth unit cell along the μ direction

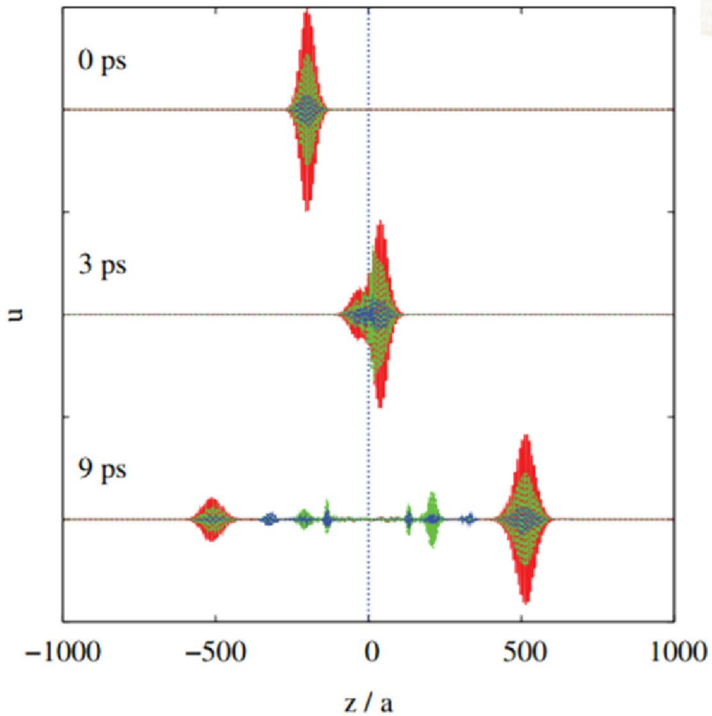
Eigenvector of polarization λ at wave vector center k_0

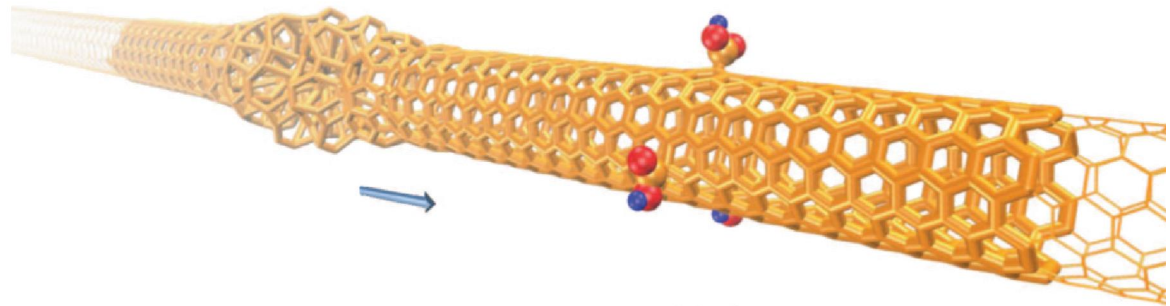
$$u_{il\mu} = A\varepsilon_{i\mu\lambda}(k_0)\exp[ik_0(z_l - z_0)]\exp[-(z_1 - z_0)^2/\eta^2]$$

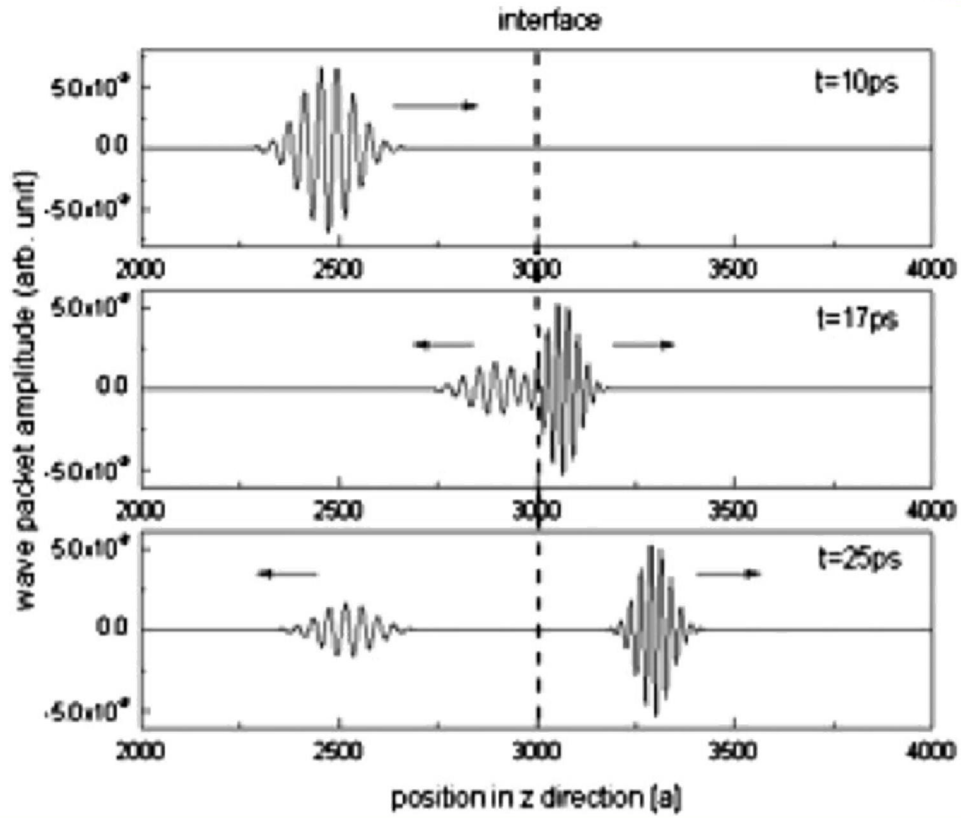
Amplitude of the wave-packet

Width of the wave-packet

Phonon Wave-packet Method







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