Mechanical Engineering

Carnegie Mellon

PREDICTING PHONON PROPERTIES FROM *AB-INITIO* MOLECULAR DYNAMICS AND NORMAL MODE DECOMPOSITION

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Supported by AFSOR FA95501010098 (2010 YIP)

Background

- Thermal conductivity of Si (dielectric) is determined by phonons:

$$\kappa = \frac{1}{V}\sum_i c_v(\omega_i) v_g^2(\omega_i)\tau_i(\omega_i)$$

- Classical molecular dynamics simulation over-predicts experiment at 300K:

$$\kappa_{SW} \sim 400\text{W/m}-\text{K}$$

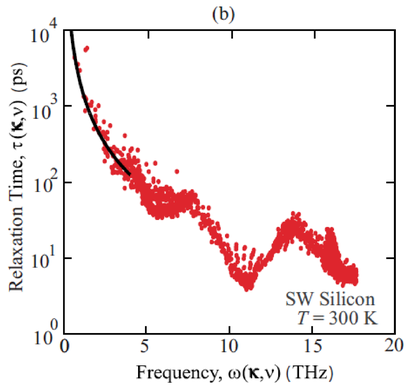
$$\kappa_{EXP} \sim 130\text{W/m}-\text{K}$$

- *Ab-Initio* molecular dynamics can model Silicon accurately to measure phonon properties:

$$\omega_i, \tau_i(\omega_i), v_g(\omega_i)$$

-Phonon lifetime/mean free path:

$$\Lambda_i(\omega_i) = \tau_i(\omega_i)\,|\,v_g(\omega_i)\,|$$



[1]E. S. Landry, Ph.D. Thesis, Carnegie Mellon University 2009

Normal Mode Decomposition

- Atomic motion is a superposition of normal modes:

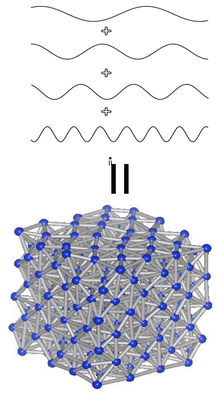
$$\dot{u}_\alpha\begin{pmatrix} l \\ b \end{pmatrix};t\Big) = \frac{1}{\sqrt{Nm_b}}\sum_{\kappa',\nu} e\begin{pmatrix} \kappa' b \\ \nu\alpha \end{pmatrix}\exp\left[i\boldsymbol{\kappa}\cdot\mathbf{r}_0\begin{pmatrix} l \\ 0 \end{pmatrix}\right]\dot{q}\begin{pmatrix} \kappa' \\ \nu \end{pmatrix};t\Big)$$

NEED:

-Equilibrium positions

- Eigenvectors from Lattice Dynamics

- Velocities from Molecular Dynamics

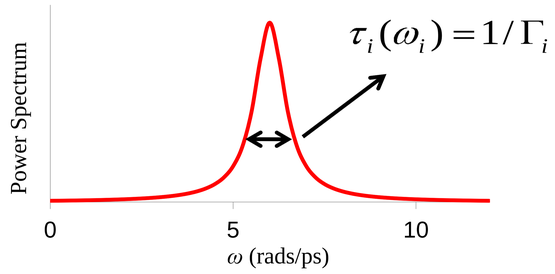


- Map atomic motion onto normal modes:

$$\dot{q}\begin{pmatrix} \kappa \\ \nu \end{pmatrix};t\Big) = \sum_{\alpha,b,l}^{3,n,N}\sqrt{\frac{m_b}{N}}\dot{u}_\alpha\begin{pmatrix} l \\ b \end{pmatrix};t\Big)e^*\begin{pmatrix} \kappa b \\ \nu\alpha \end{pmatrix}\exp\left[i\boldsymbol{\kappa}\cdot\mathbf{r}_0\begin{pmatrix} l \\ 0 \end{pmatrix}\right]$$

- Power spectrum of normal modes:

$$\Phi\begin{pmatrix} \kappa \\ \nu \end{pmatrix};\omega\Big) = \left|\int_{-\infty}^{\infty}\dot{q}\begin{pmatrix} \kappa \\ \nu \end{pmatrix};t\Big)\exp(i\omega t)dt\right|^2$$



Normal Mode Spectral Energy is Lorentzian with peak at phonon frequency.

Ab-Initio Molecular Dynamics

Density Functional Theory:

$$\hat{H}\Psi = \left[\sum_i^N -\frac{\hbar^2}{2m}\nabla_i^2 + \sum_i^N V(\vec{r}_i) + \sum_{i<j}^N U(\vec{r}_i,\vec{r}_j)\right]\Psi = E\Psi$$

Molecular Dynamics:

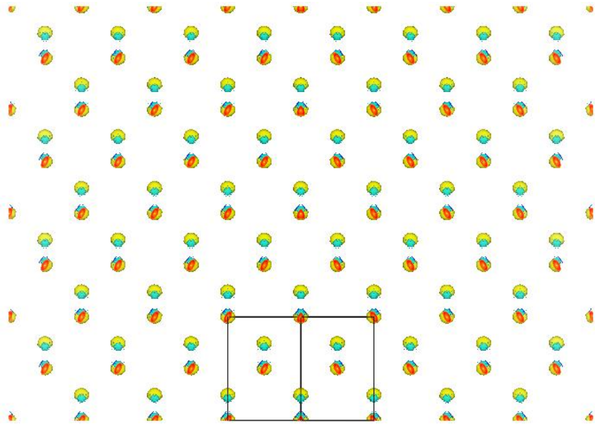
$$\vec{F} = m\vec{a} = -\nabla E = \left\langle \psi \,|\, \frac{\partial \hat{H}}{\partial \vec{R}} \,|\, \psi \right\rangle$$

CP2K[2]:

- Used 128 CPU on Diamond and Garnet

- Mixed Gaussian and Plane Wave code:

$$\psi_{nk}(\vec{r}) \propto \exp(-\alpha\vec{r}^{\,2}) \propto \sum_k C_k \exp(i\vec{k}\cdot\vec{r})$$



Si electronic kinetic energy density calculated using Density Functional Theory

[2]http://cp2k.berlios.de/

Simulation Expense

CP2K

• 3 steps/min/128cpu =

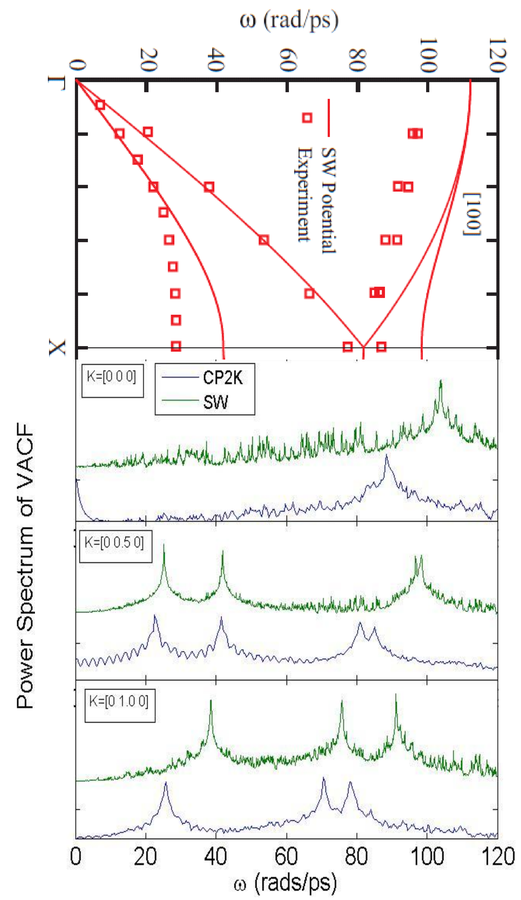
0.023 steps/min/cpu

Stillinger-Weber

•6E4 steps/min/4 cpu =

15000 steps/min/cpu

Power Spectrum



Vibrational power spectrum for Si from CP2K and Stillinger-Weber with matching dispersion curve.