Predicting Phonon Properties of Defected Systems using the Spectral Energy Density

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Dielectric Thermal Conductivity

- **Dielectric crystal = Electrical Insulator**

- Ex: Si, Ge

$$\vec{q} = -\kappa \nabla T$$

- **Dielectric Thermal Conductivity:**

$$\kappa_{total} = \kappa_{phonon} \cancel{+ \kappa_{elec}}$$

- **Phonons** are lattice vibrations…

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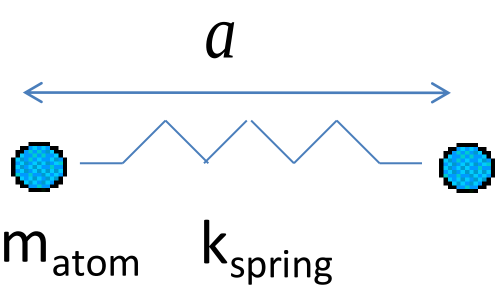
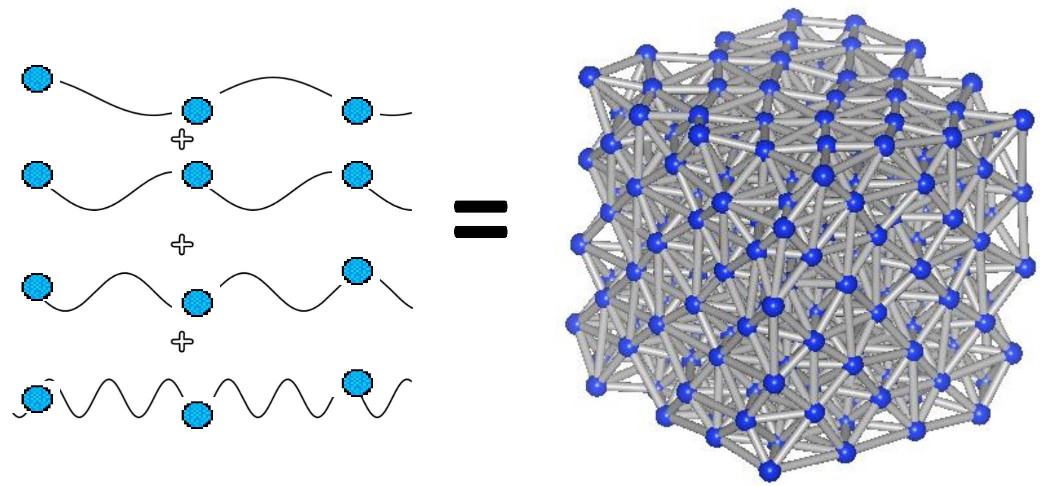
Phonons

<u>Lattice vibrations (Phonons) are travelling waves:</u>

$$u(x) \propto e^{i(kx-\omega t)} \longrightarrow v_g$$





$$F \propto kx + \varepsilon x^3 + \ldots$$

<u>Phonons Interact:</u>

- Non-linear springs cause phonons to interact.

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Thermal Conductivity Phonon Gas

Kinetic Theory Phonon Gas:

- If system L >> Λ :

$$\kappa = \frac{1}{3}\rho c_v v_g \Lambda$$

-**Phonons interact**: with each other, defects, boundaries, etc.

$$\kappa = \frac{1}{3}\rho c_v v_g \Lambda \quad \Rightarrow \quad \kappa = \frac{1}{V}\sum_i c_v(\omega_i) v_g^2(\omega_i)\tau_i(\omega_i)$$

Phonons interact

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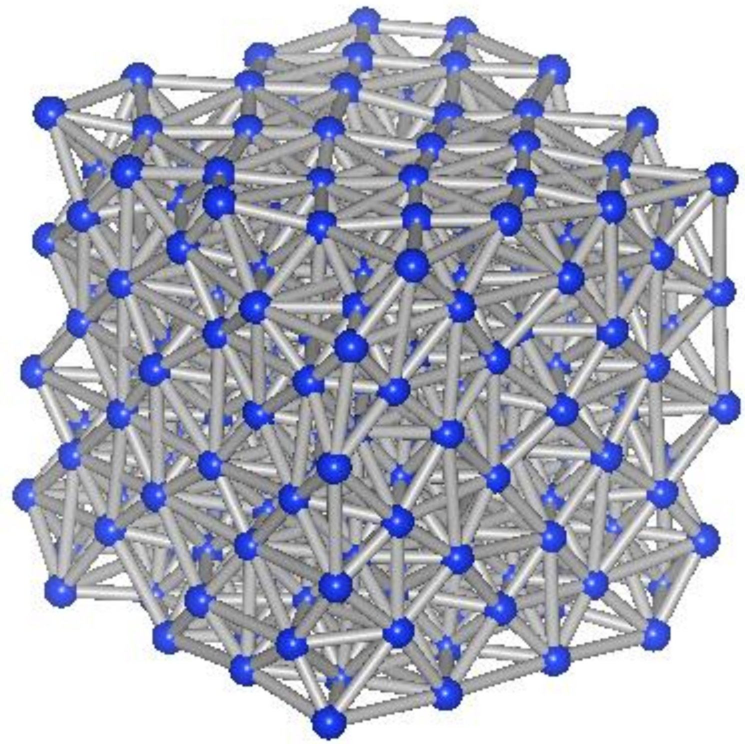
Molecular Dynamics and Spectral Energy Density

- **Molecular Dynamics**

- Lennard-Jones:

$$V(r) = 4\varepsilon\left(\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right)$$



- **Spectral Energy Density**

- **Frequency, group velocity** and **lifetimes** of phonons from Molecular Dynamics.

$$\vec{r}(t), \vec{p}(t) \longrightarrow \omega_i, \tau_i(\omega_i), v_g(\omega_i)$$

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Spectral Energy Density

$$\Phi(\omega,\boldsymbol{\kappa}) = 2\sum_{\nu}^{3n}\sum_{\alpha,b}^{3,n}\left\langle T\left(\begin{smallmatrix}\boldsymbol{\kappa} & b \\ \nu & \alpha\end{smallmatrix};\omega\right)\right\rangle$$

Spatial and temporal fourier txn.

$$= \frac{1}{4\pi\tau_0 N}\sum_{b}^{n} m_b \sum_{\alpha}^{3}\left|\int_{-\tau_0}^{\tau_0}\sum_{l}^{N}\dot{u}_\alpha\left(\begin{smallmatrix}l \\ b\end{smallmatrix};t\right)\exp\left[i\boldsymbol{\kappa}\cdot\mathbf{r}_0\left(\begin{smallmatrix}l \\ 0\end{smallmatrix}\right) - i\omega t\right]dt\right|^2$$

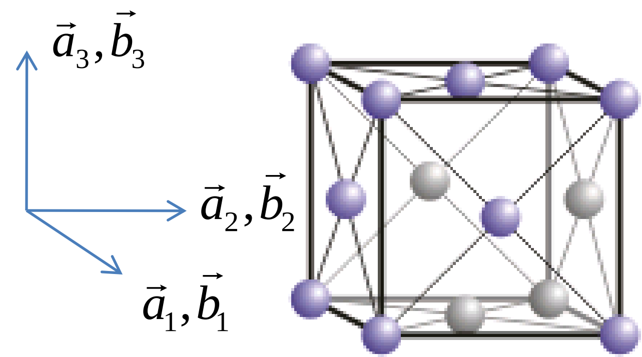
mass

velocity

allowed wavevectors and equil. positions

- **No phonon knowledge required *a priori*.**
- **Can measure:**

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



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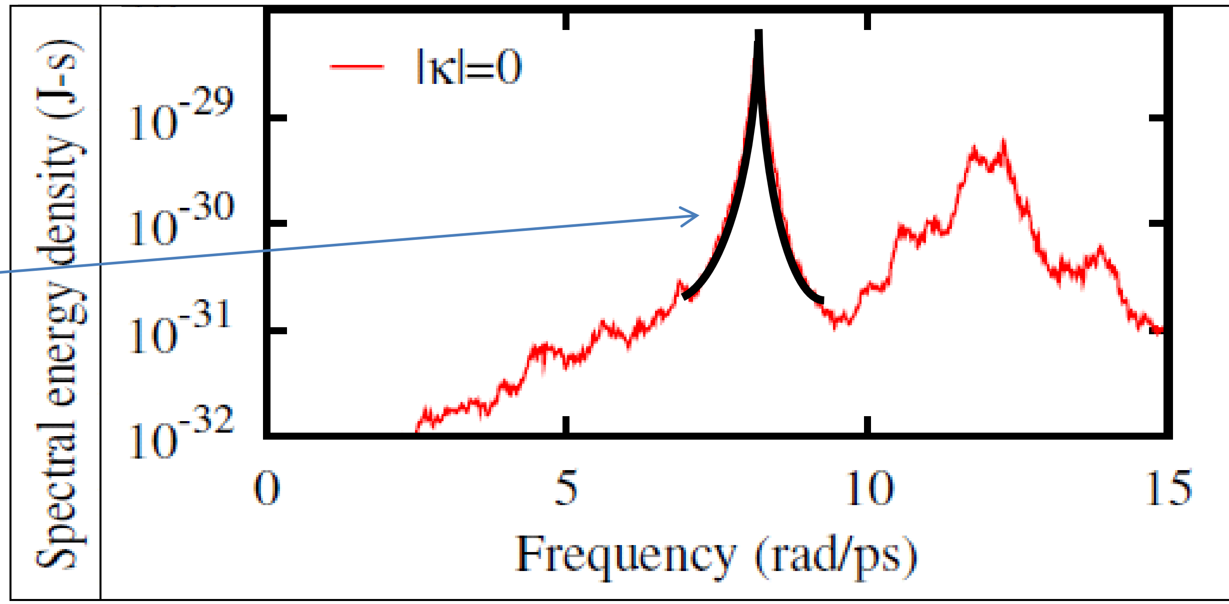
Spectral Energy Density Pure System

- **Spectral Energy Density (SED):** system energy in frequency and wavevector space.

$$\tau_i(\omega_i) = 1/2\Gamma$$



Broad peak = short **lifetime**

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

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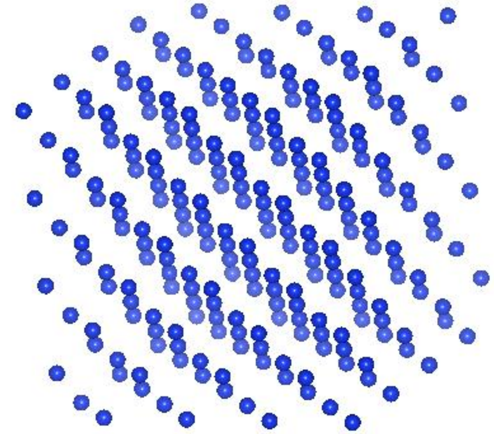
Spectral Energy Density of Defected System

Defect = Disorder = non-periodic

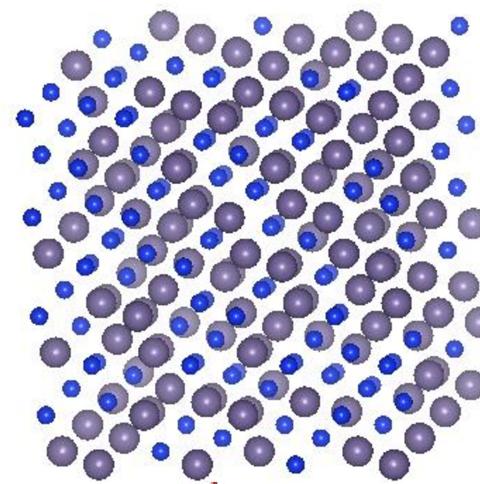
$$\mathbf{m1_{1-x}m2_x}$$

m1=1.0 **m2**=3.0

$x = 0.0$



$x = 0.5$



Phonon picture still valid?

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Bulk Thermal Conductivity Prediction

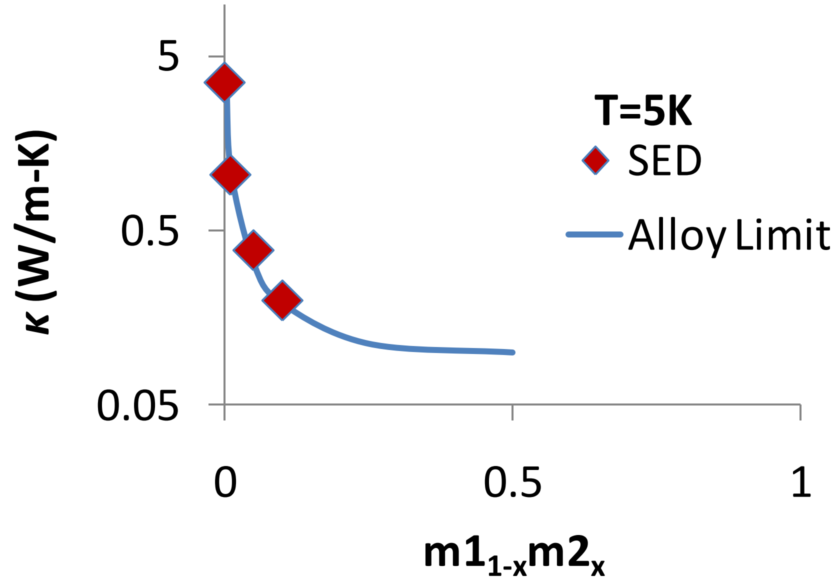
$$\kappa = \frac{1}{V}\sum_i c_v(\omega_i) v_g^2(\omega_i)\tau_i(\omega_i)$$

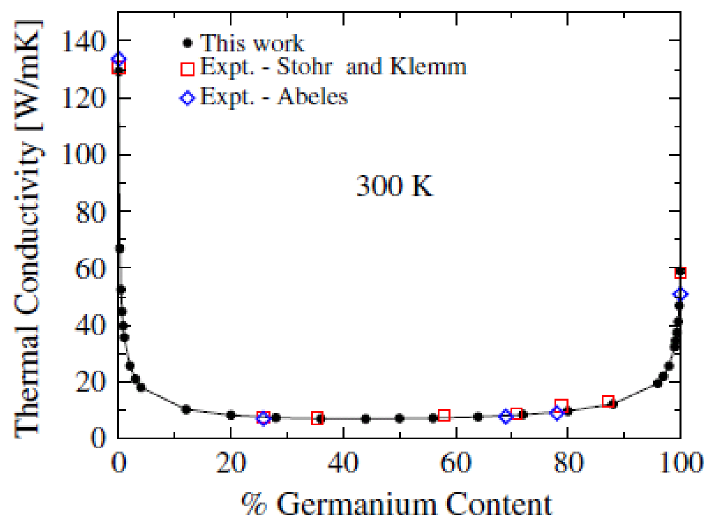
- **Predict thermal conductivity up to x=0.1 (weakly perturbed)**

- **<u>Typically:</u>**

$$0.9\kappa_{alloy}^{0.1} \approx \kappa_{alloy}^{0.5}$$

- **m1/m2 = 3 ($m_{Si}/m_{Ge}\approx 3$)**





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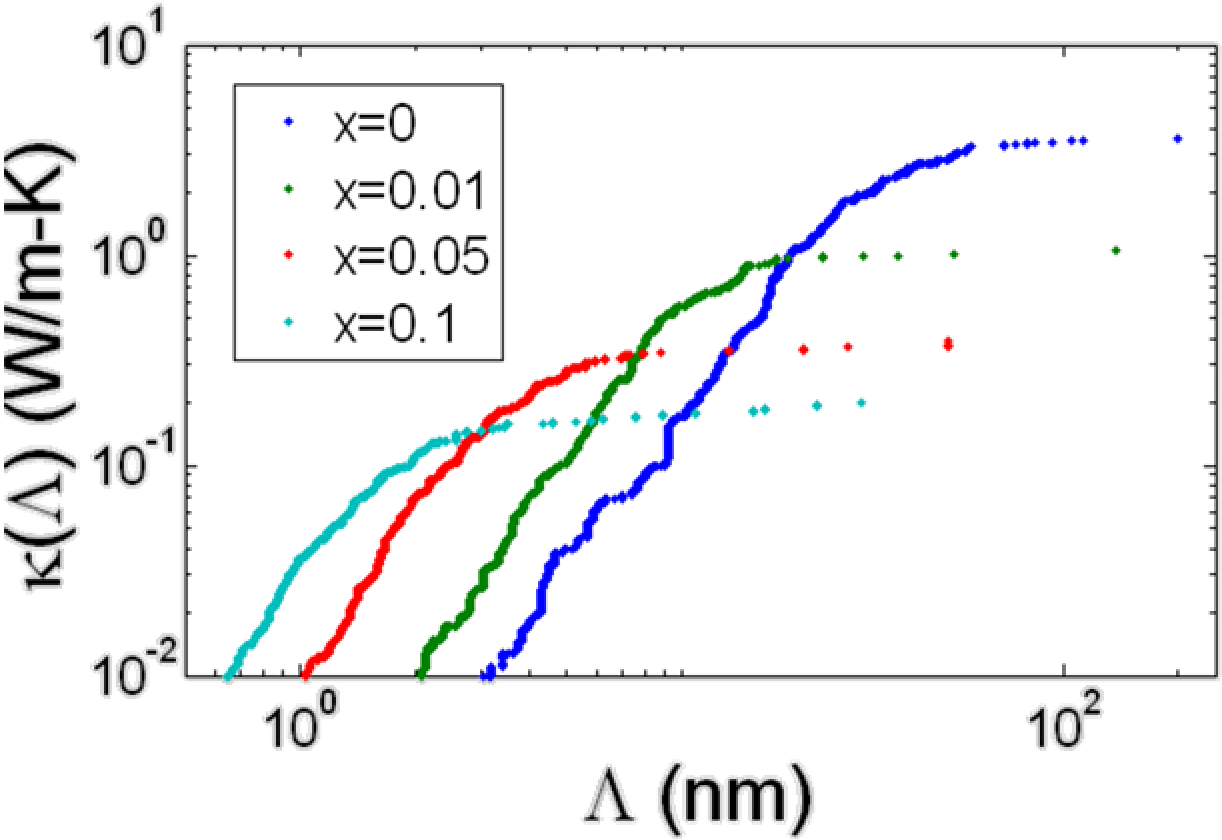
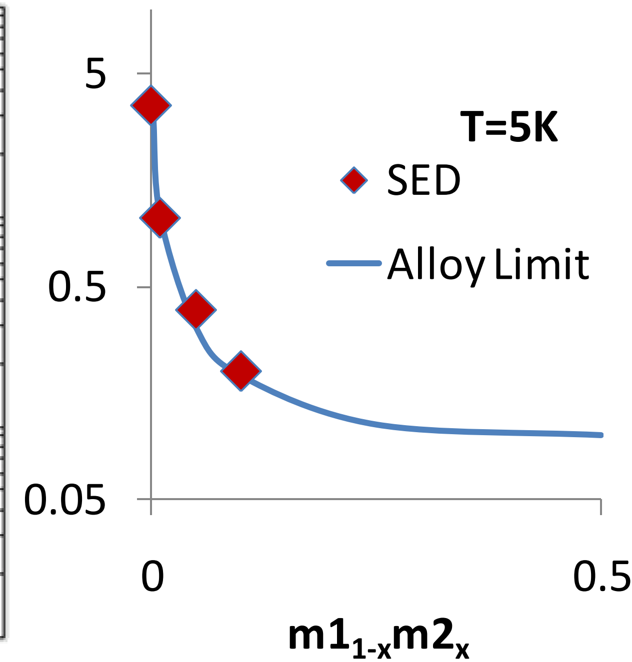
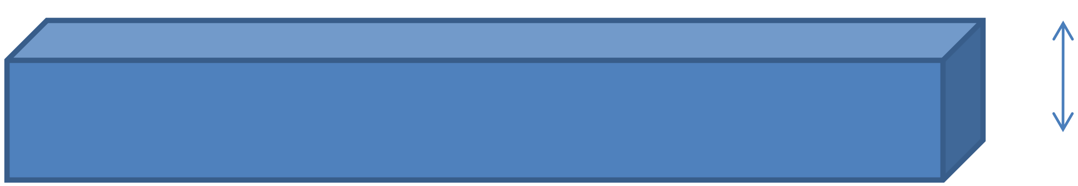
Thermal Conductivity of Thin Films





$$\kappa = \frac{1}{V}\sum_i c_v(\omega_i) v_g(\omega_i) \Lambda_i(\omega_i)$$



$$L \approx \Lambda$$

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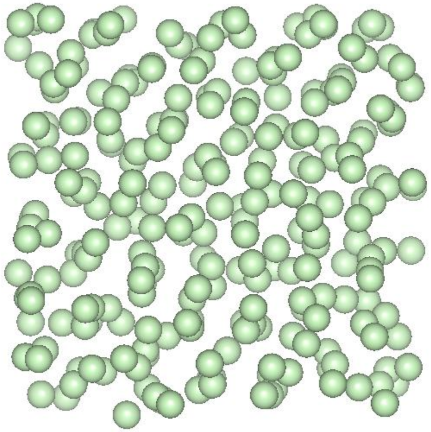
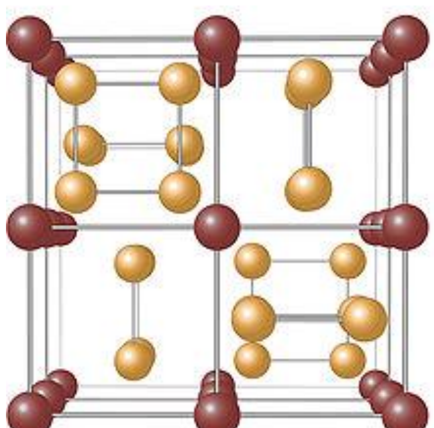
Thermal Conductivity Amorphous (Heavily Disordered) Materials



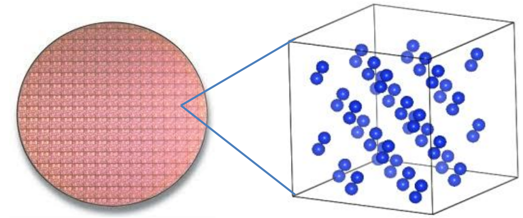
$$\kappa_{total} = \kappa_{phonon} + \kappa_{disorder}$$

$\vec{k}, v_g$

$\vec{k}, \cancel{v_g}$

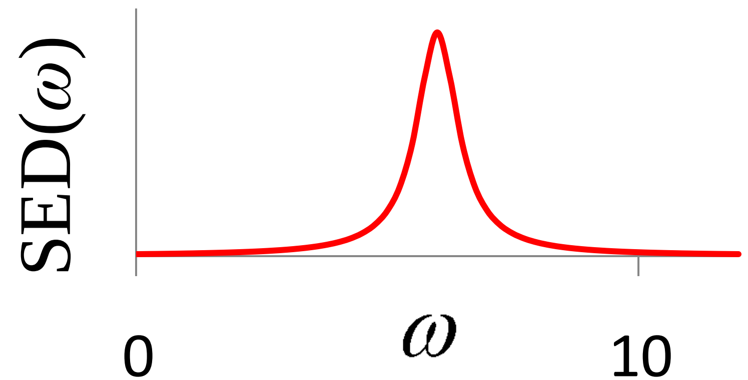


Discussion



- Dielectric thermal conductivity can be described by Kinetic Theory (bulk system).

$$\vec{q} = -\kappa \nabla T$$



- Molecular Dynamics and Spectral Energy Density can measure phonon properties.

- Phonon properties can be predicted for "weakly" perturbed systems, analyzed on mode by mode basis.

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

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Questions

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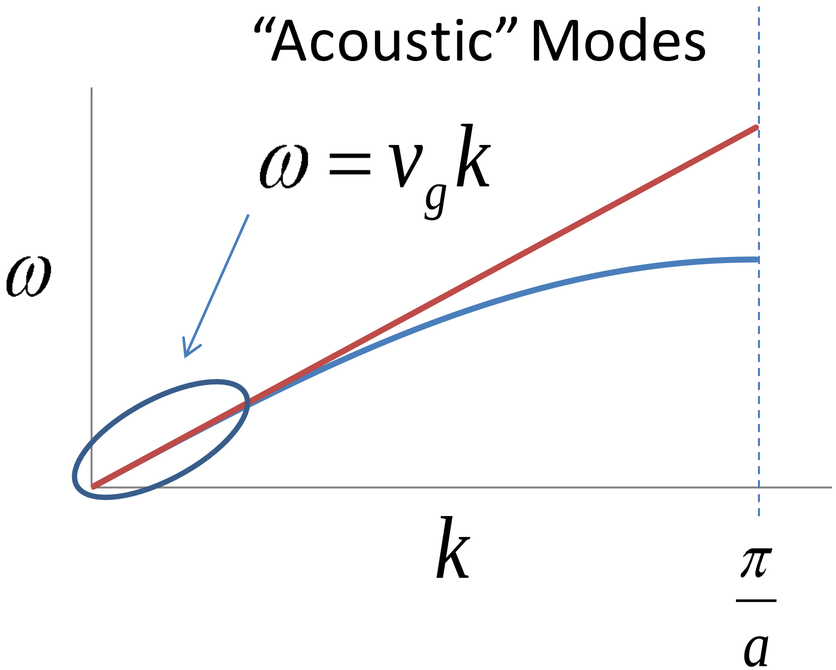
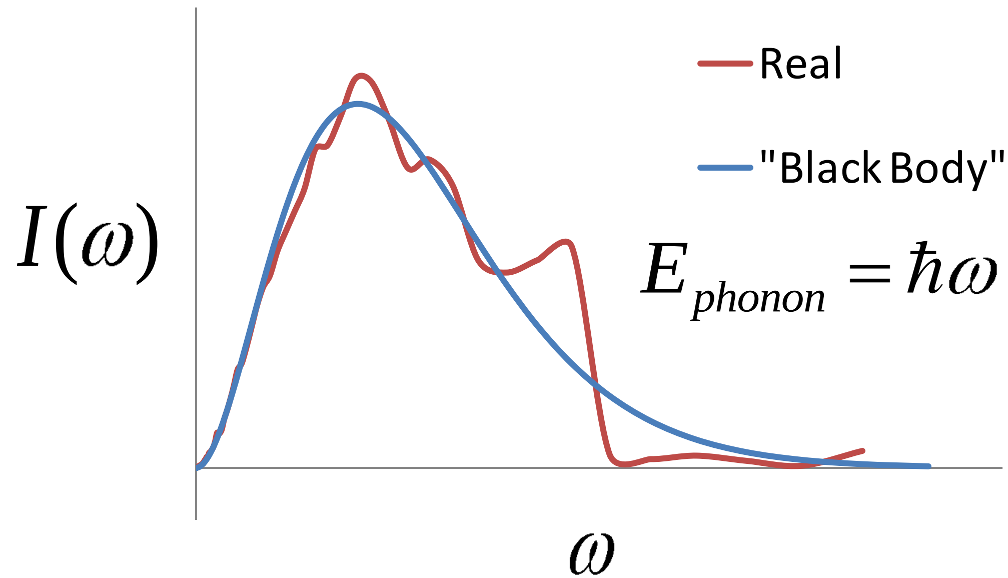
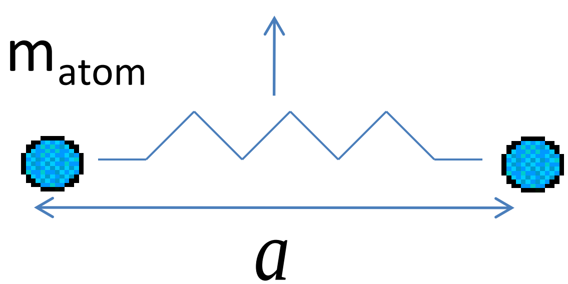
Phonon Gas





Phonons vs. Photons:

$$F \propto kx + \varepsilon x^3 + \ldots$$



- Non-linear springs
- Phonons interact with each other
- **Interacting Gas...**

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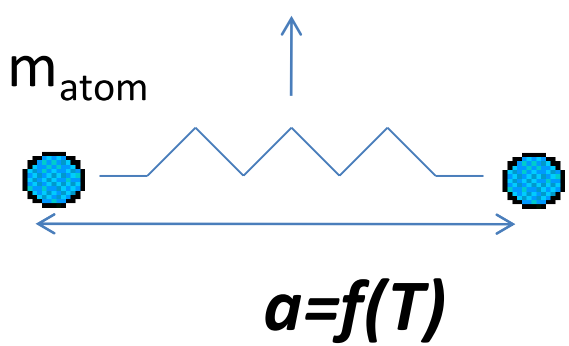
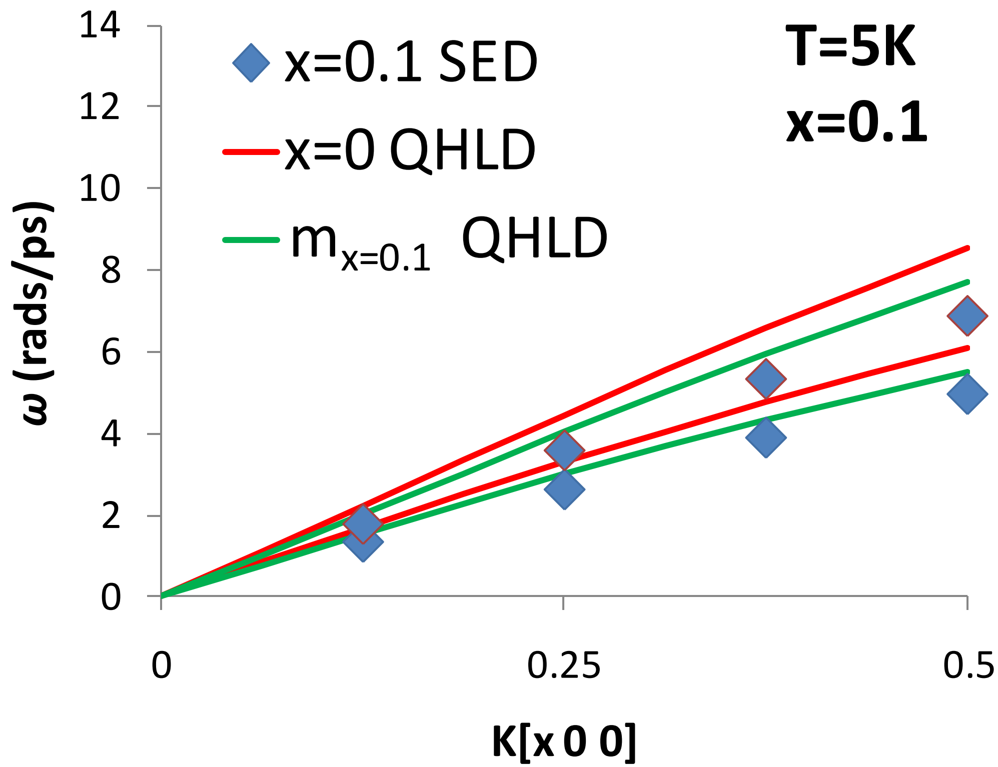
Dispersion of Disordered System

- **Quasi-Harmonic Lattice Dyanmics:**

$$F \propto kx$$





-**Virtual Crystal Approximation:**

$m_{x=0.1} = (0.9m1+0.1m2) = 1.2$

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