A Correlation Perspective

- Correlation based description
  - Liouville Eq. → Linear response theory
  - Causality → Correlation

$$\vec{\mathbf{Q}}(t) = \frac{1}{V}\sum_{j}^{N}\left[E_j \cdot \vec{\mathbf{v}}_j + \sum_{l}^{N}\frac{\partial \Phi_l}{\partial \vec{\mathbf{r}}_j}\cdot \vec{\mathbf{v}}_j \cdot \vec{\mathbf{r}}_{jl}\right] \quad \kappa = \frac{V}{k_B T^2}\int_0^{\infty}\left\langle \vec{\mathbf{Q}}(t)\cdot \vec{\mathbf{Q}}(t+t')\right\rangle dt'$$

  - Green-Kubo relations
  - General: Valid for any phase of matter!
- Mode-mode interactions
  - PGM: scattering → impedes heat flow
  - GK: correlation → facilitates heat flow

Modal Analysis

- Green-Kubo Modal Analysis (GKMA)

$$\dot{X}_n(t) = \sum_j \dot{\vec{\mathbf{X}}}_j(t) \cdot \vec{\mathbf{p}}_j(n) \qquad \dot{\vec{\mathbf{X}}}_j(t) = \sum_n \dot{X}_n(t) \cdot \vec{\mathbf{p}}_j(n) = \boxed{\sum_n \vec{\mathbf{v}}_j(n,t)}$$

$$\vec{\mathbf{Q}}(t) = \sum_n \frac{1}{V} \sum_j^N \left[E_j \cdot \vec{\mathbf{v}}_j(n,t) + \sum_l^N \frac{\partial \Phi_l}{\partial \vec{\mathbf{r}}_j} \cdot \vec{\mathbf{v}}_j(n,t) \cdot \vec{\mathbf{r}}_{jl} \right] = \sum_n \vec{\mathbf{Q}}(n,t)$$

$$\kappa = \sum_n \sum_{n'} \frac{V}{k_B T^2} \int_0^\infty \left\langle \vec{\mathbf{Q}}(n,t) \cdot \vec{\mathbf{Q}}(n',t+t') \right\rangle dt' = \boxed{\sum_n \sum_{n'} \kappa(n,n')}$$

- Interface Conductance Modal Analysis (ICMA)

$$Q(t) = \sum_n \sum_{j,l}^N \vec{\mathbf{F}}_{jl} \cdot \left(\vec{\mathbf{v}}_j(n,t) + \vec{\mathbf{v}}_l(n,t) \right) = \sum_n Q(n,t)$$

$$G = \sum_n \sum_{n'} \frac{1}{k_B T^2} \int_0^\infty \left\langle Q(n,t) \cdot Q(n',t+t') \right\rangle dt' = \boxed{\sum_n \sum_{n'} G(n,n')}$$

GKMA & ICMA Implementation

- Need to multiply $\vec{\mathbf{F}}_{jl} \cdot \vec{\mathbf{v}}_j(n,t)$

$$\vec{\mathbf{Q}}(t) = \sum_n \frac{1}{V} \sum_j^N \left[E_j \cdot \vec{\mathbf{v}}_j(n,t) + \sum_l^N \frac{\partial \Phi_l}{\partial \vec{\mathbf{r}}_j} \cdot \vec{\mathbf{v}}_j(n,t) \cdot \vec{\mathbf{r}}_{jl} \right]$$

$$Q(t) = \sum_n \sum_{j,l}^N \vec{\mathbf{F}}_{jl} \cdot \left(\vec{\mathbf{v}}_j(n,t) + \vec{\mathbf{v}}_l(n,t) \right)$$

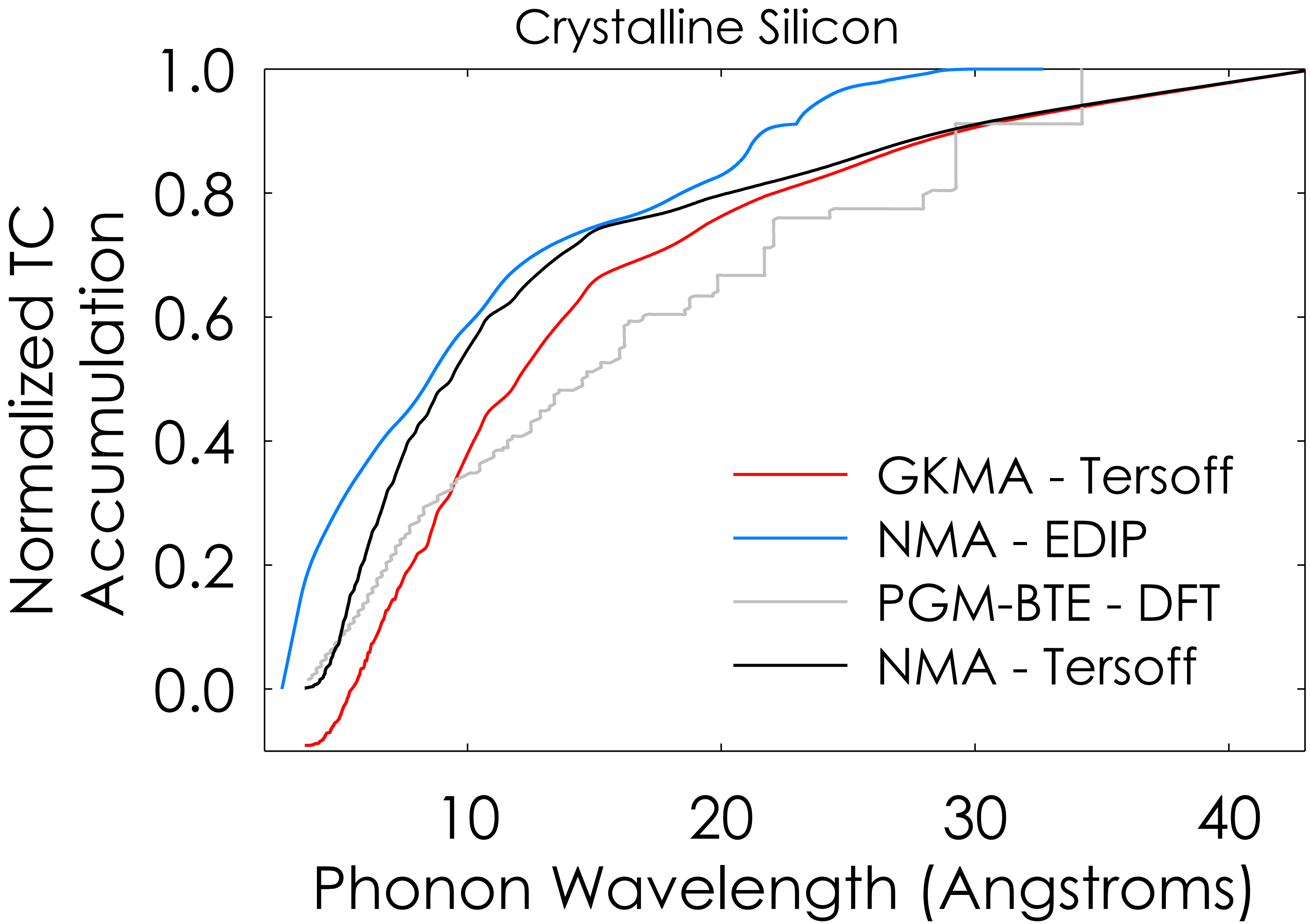
- Implement code inside force routine
- LAMMPS
- Decompose trajectory
  - Break into frequency bins
- Multiply modal velocity components
  - Determine mode heat flux

Quantum Correction

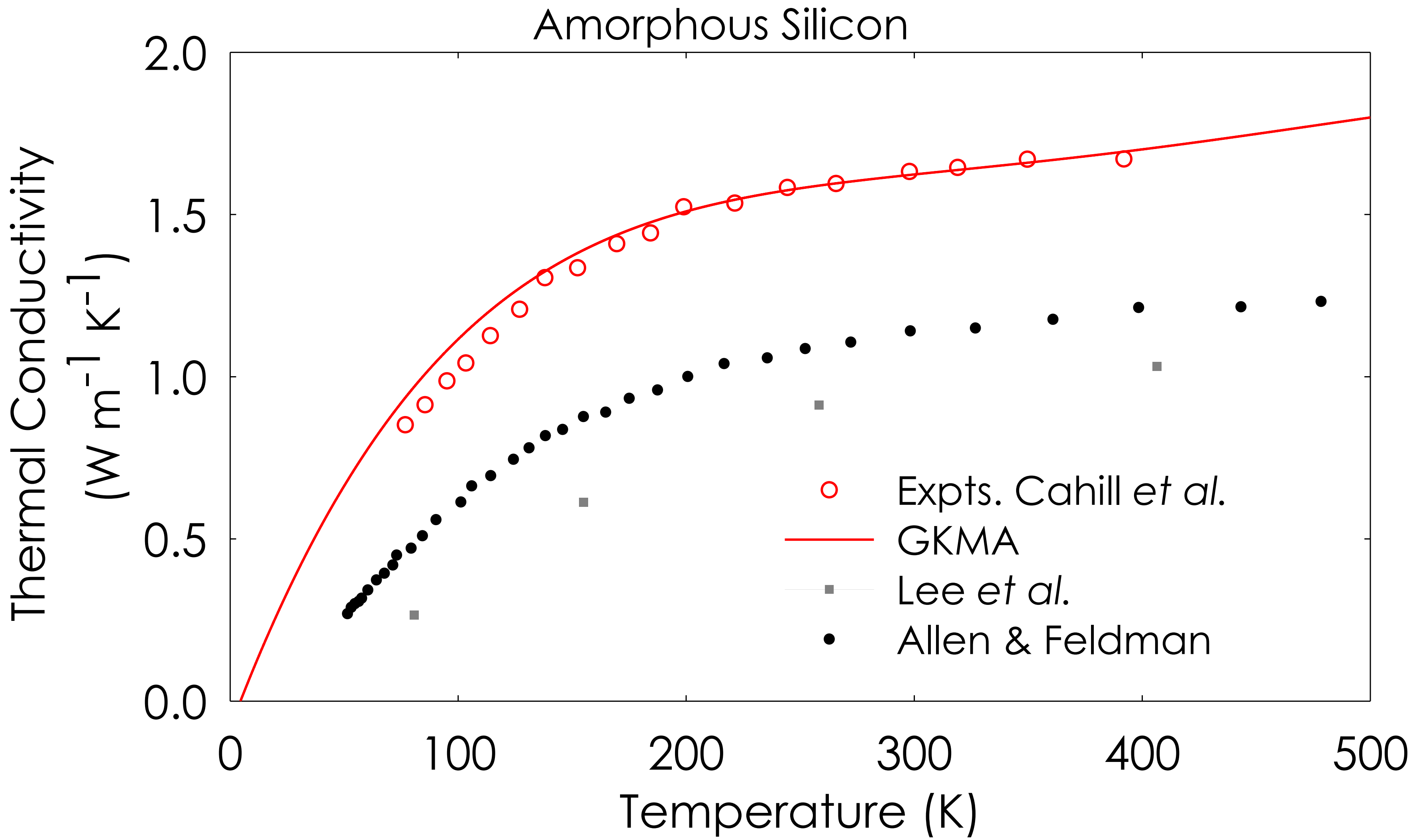
- Contributions are proportional to C_p
- Classical mode amplitudes incorrect
  - Fully excited
  - Constant C_p vs. T
  - Naturally result from MD
- Quantum mode amplitudes correct
  - Strong temperature dependence
  - High T limit = classical
  - Closed form expression
  - Correction factor

$$C_p = \sum_n k_B \left[x^2 \left(e^x - 1 \right)^2 \right] \qquad x = \frac{\hbar\omega}{k_B T}$$

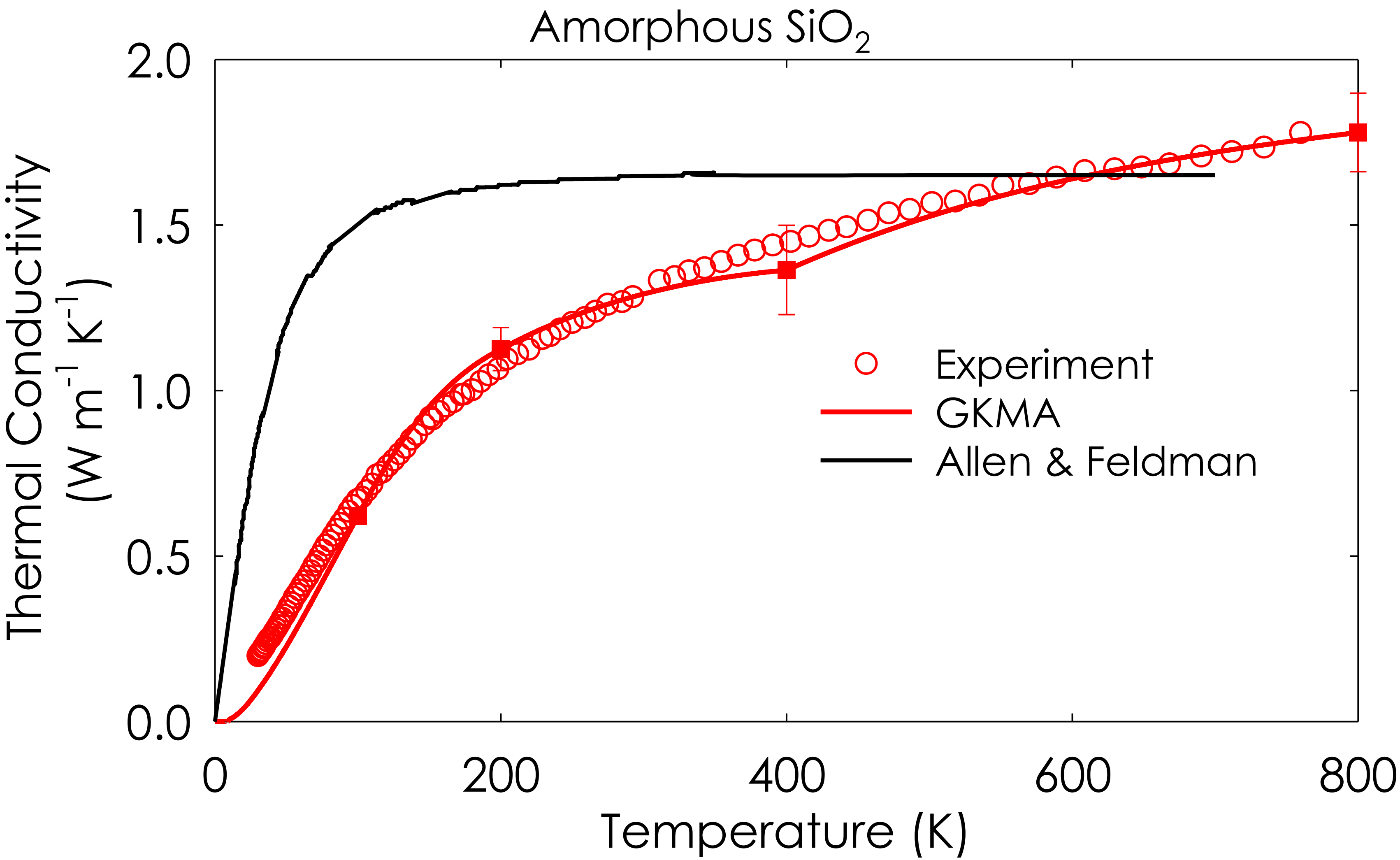
GKMA Validation



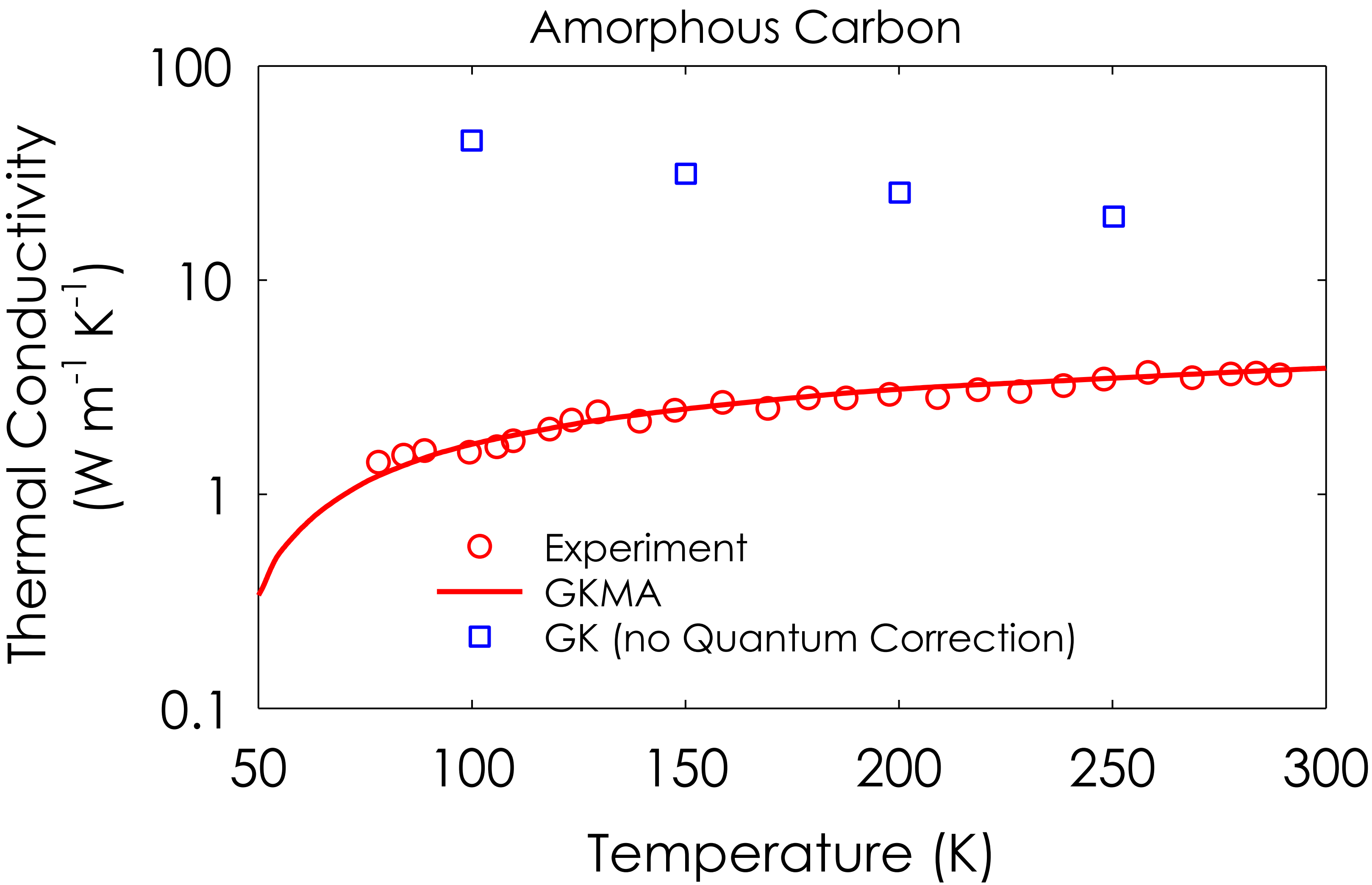
Testing & Validation



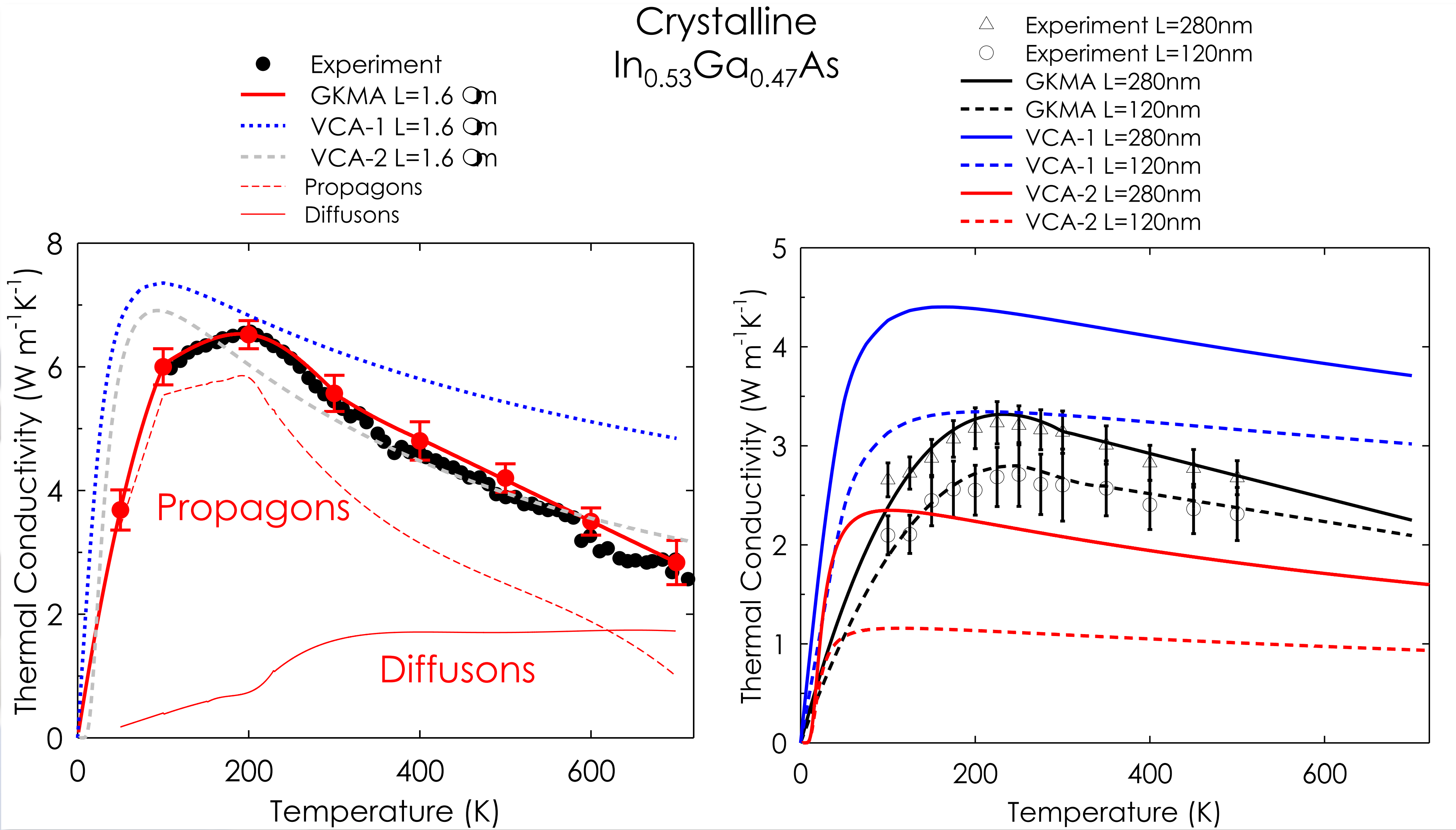
Testing & Validation



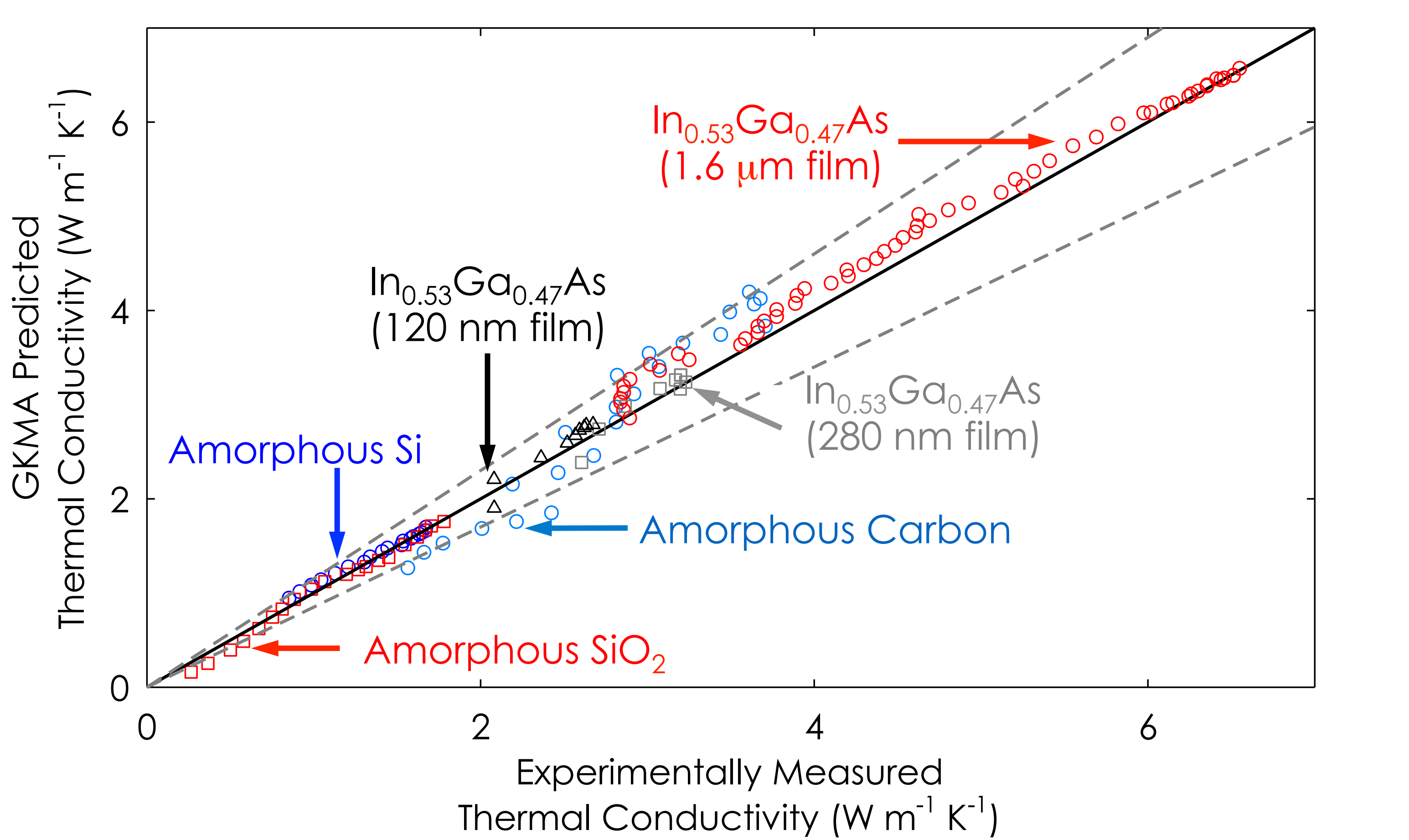
Testing & Validation



Testing & Validation

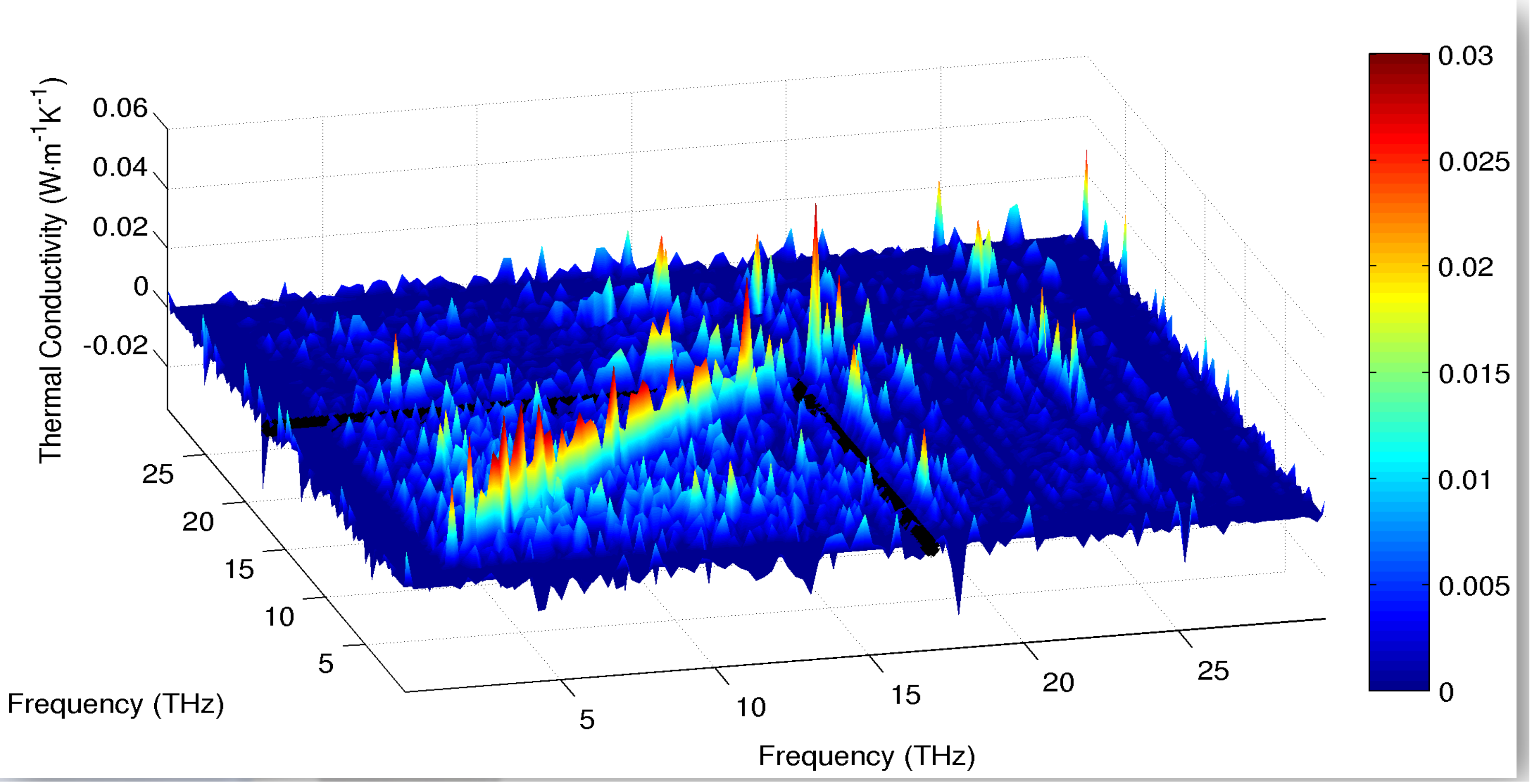


GKMA Validation

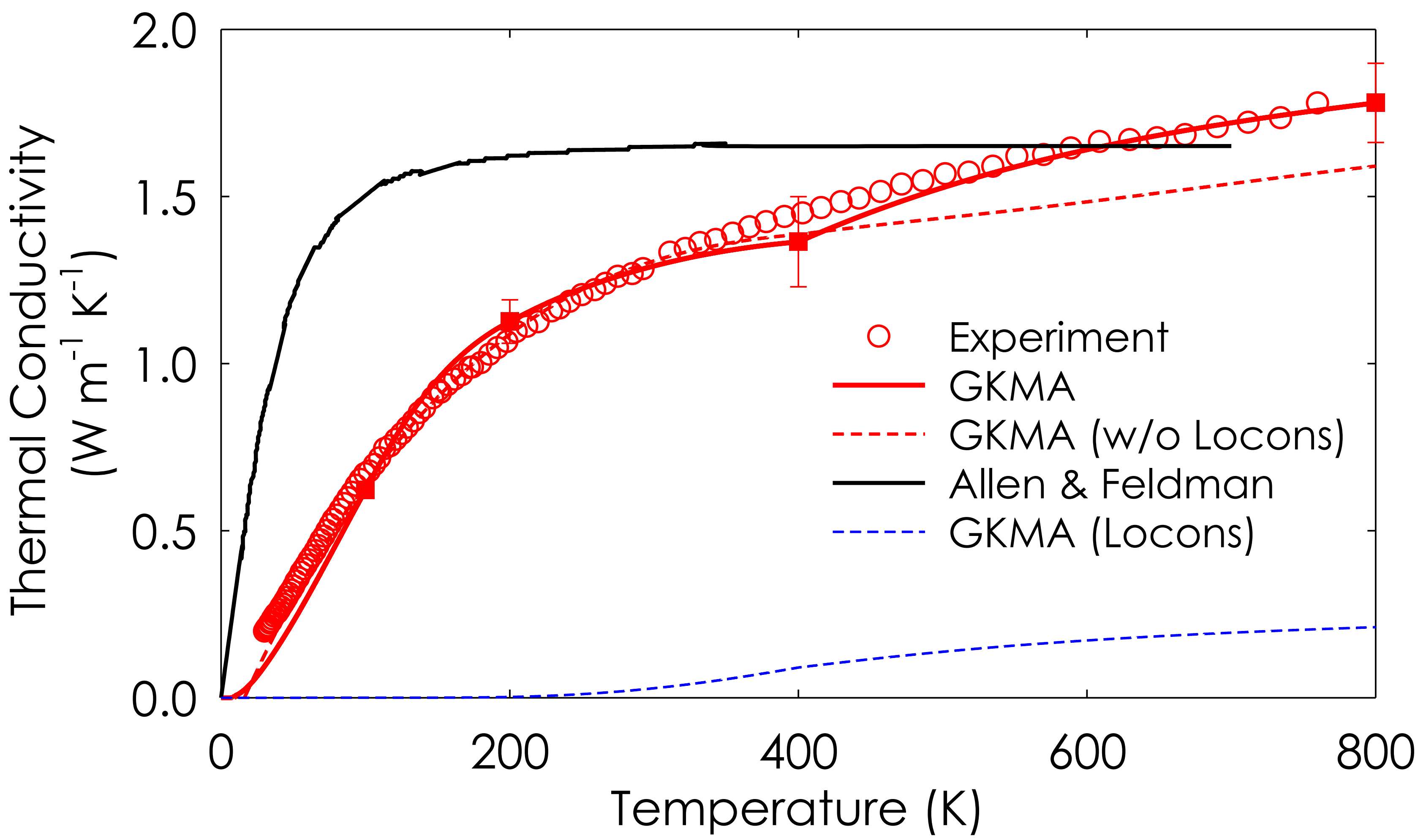


Correlation Maps (a-Si)

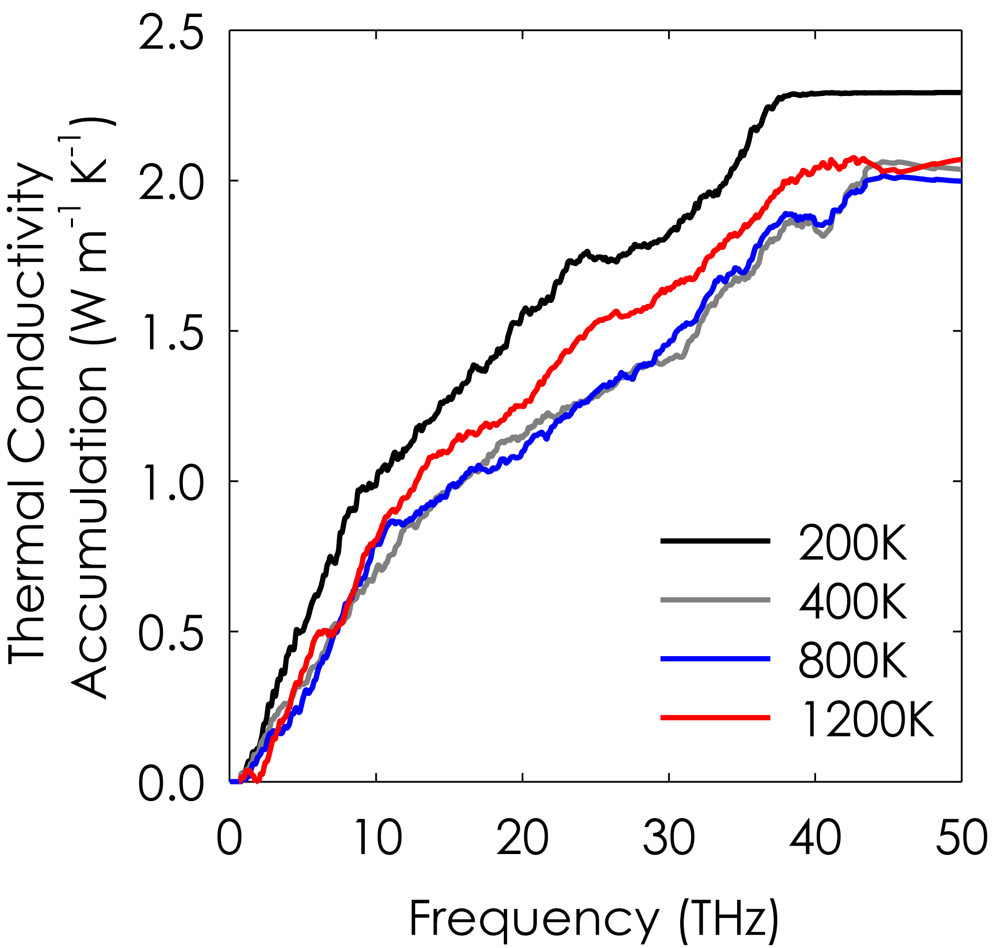
$$\kappa_n \propto \int \langle Q_n(t) \cdot Q_{total}(t+t') \rangle dt' = \sum_{n'} \int \langle Q_n(t) \cdot Q_{n'}(t+t') \rangle dt'$$

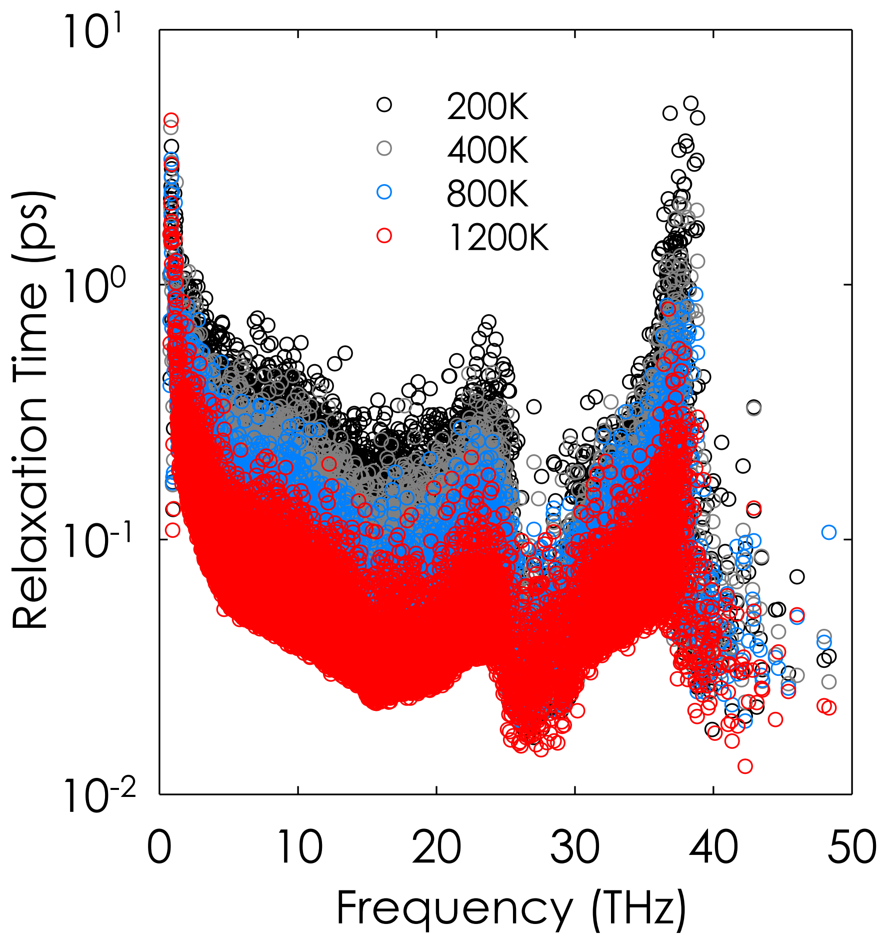


Locons Can Contribute!

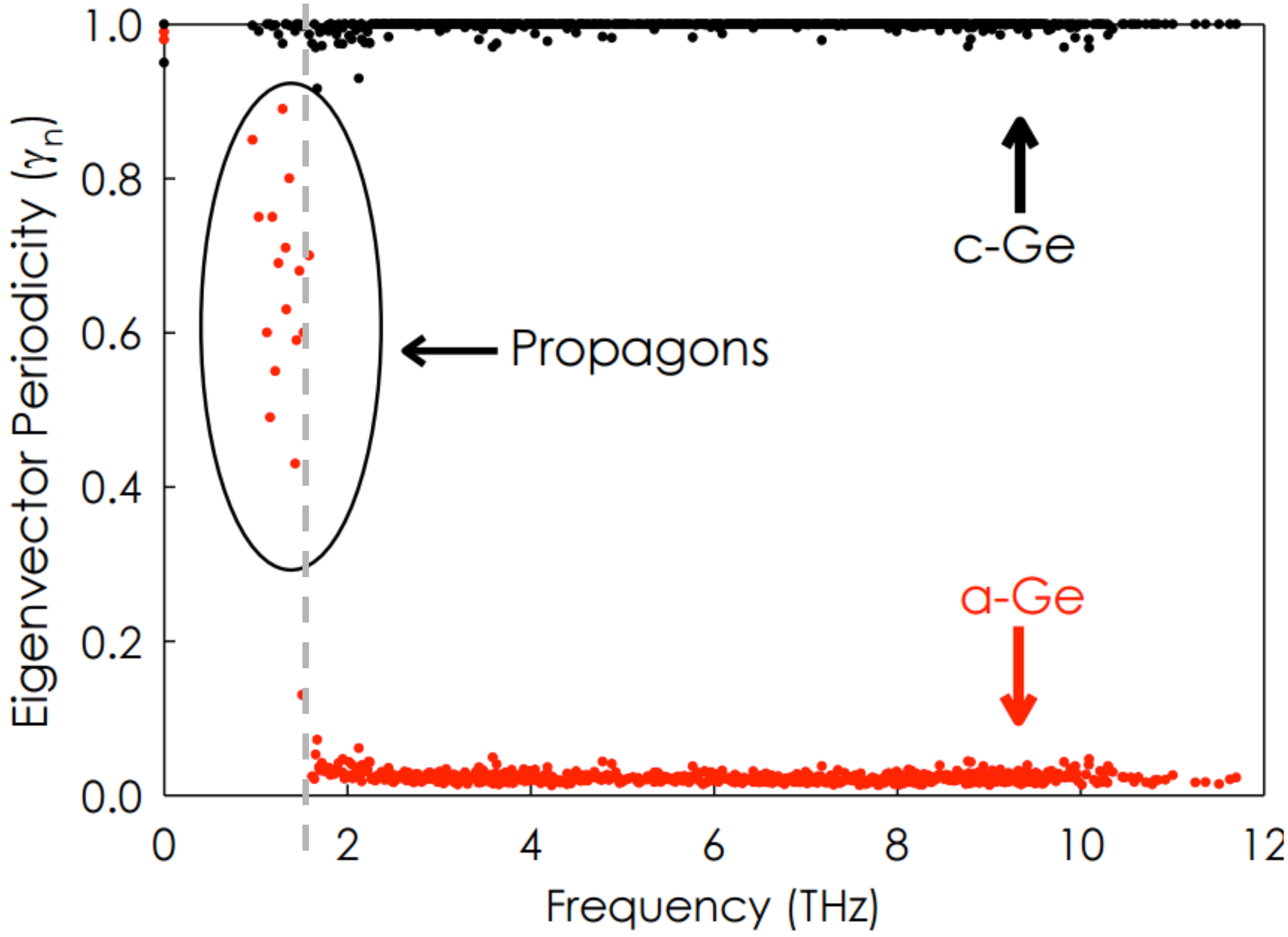


Relaxation Time → Not A Descriptor

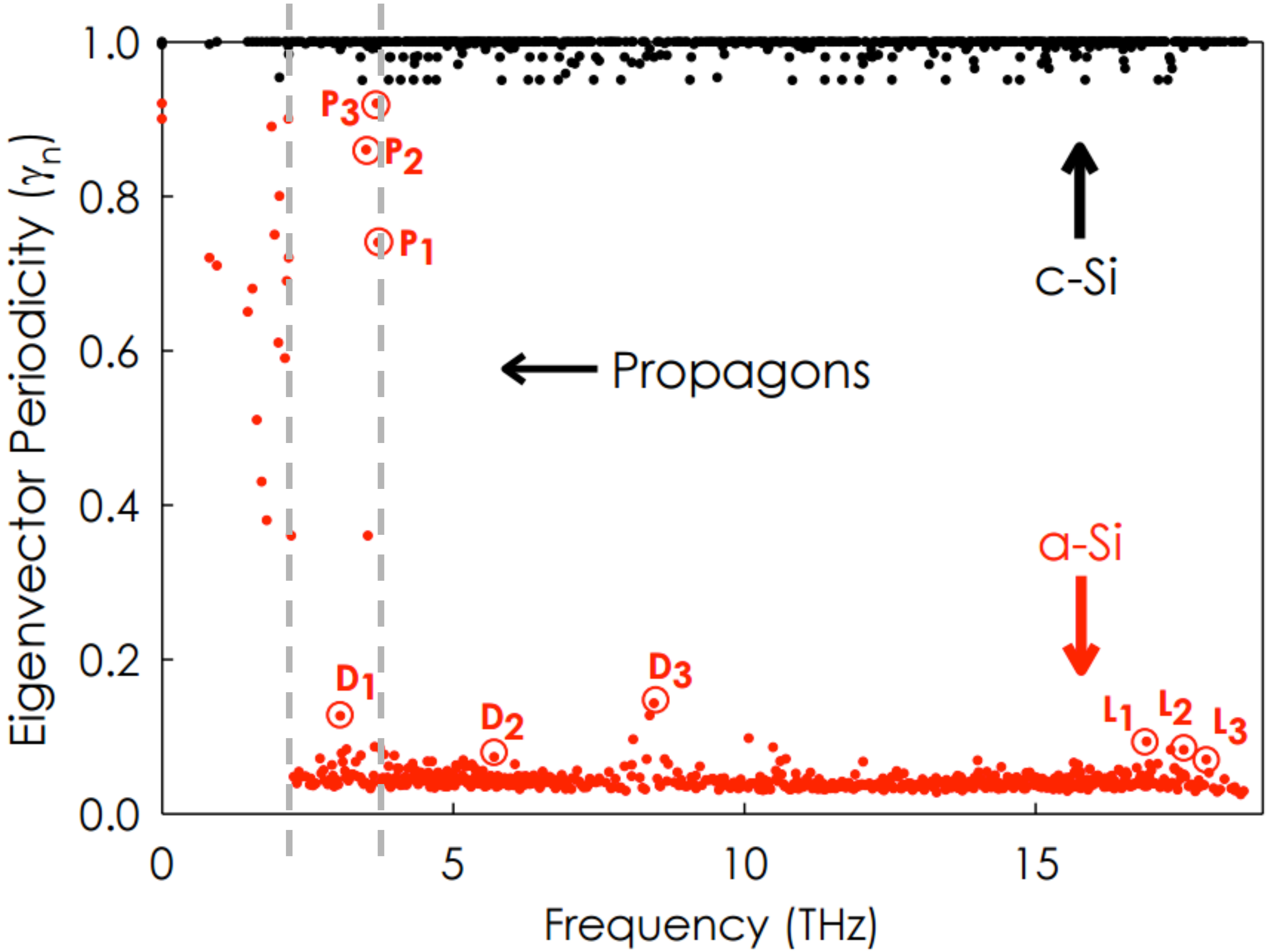




We Can Distinguish Between the Modes

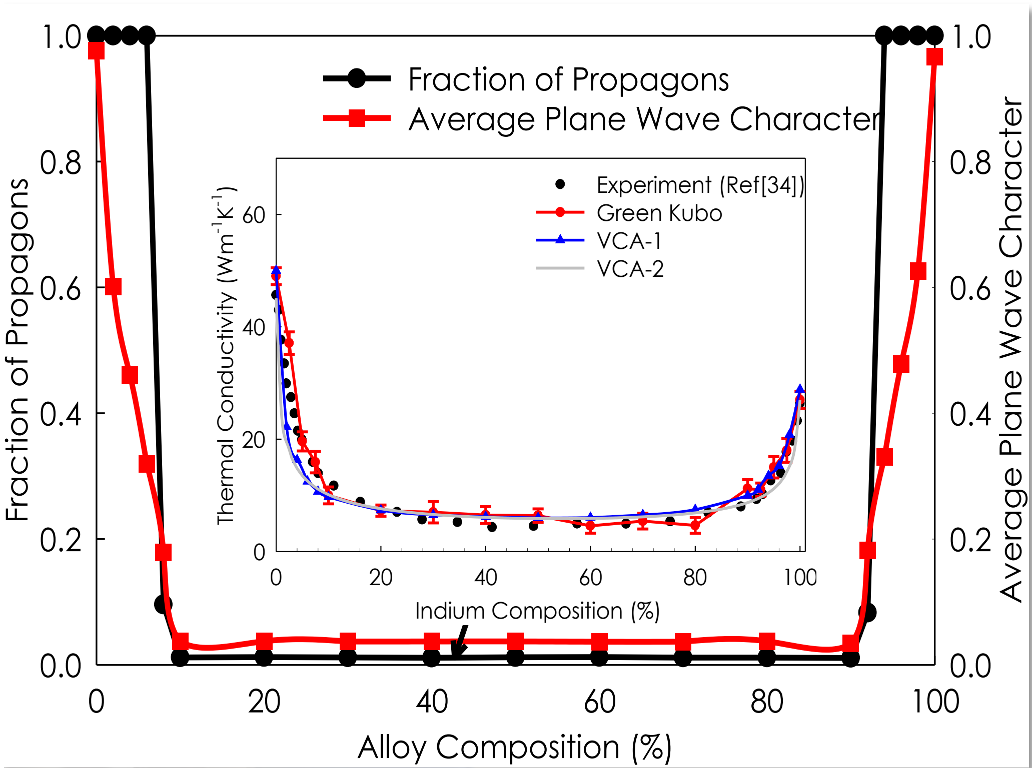


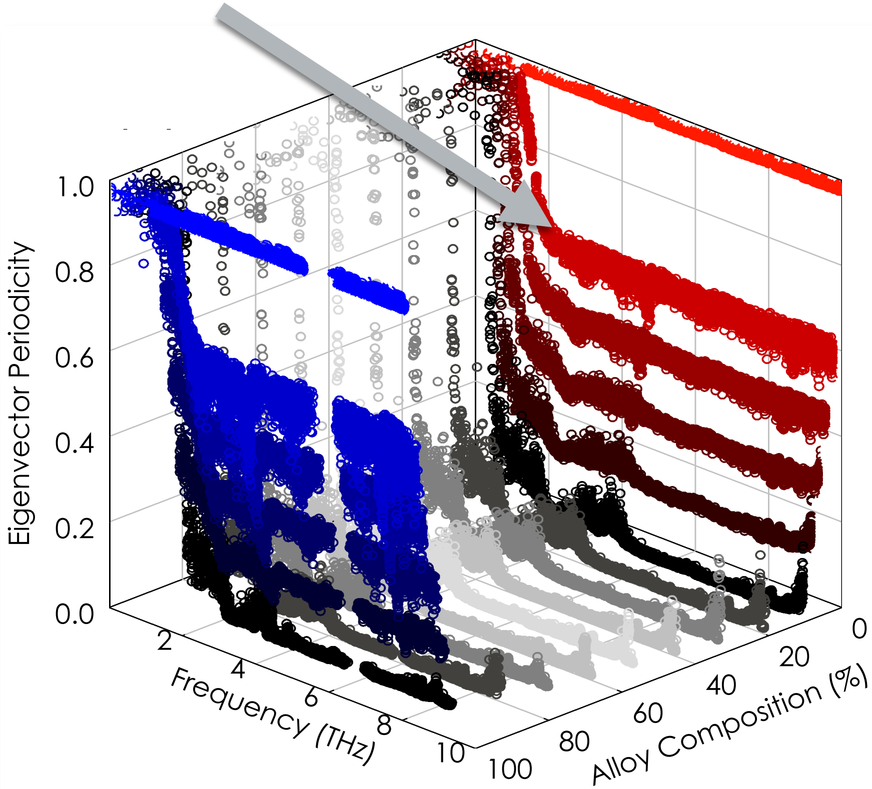
We Can Distinguish Between the Modes



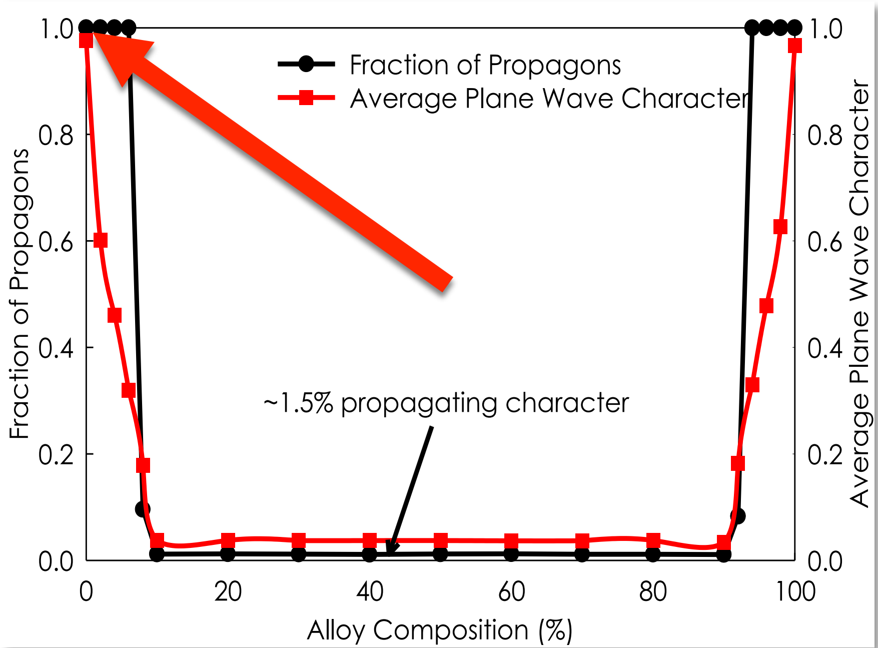
Mode Character Changes Quickly

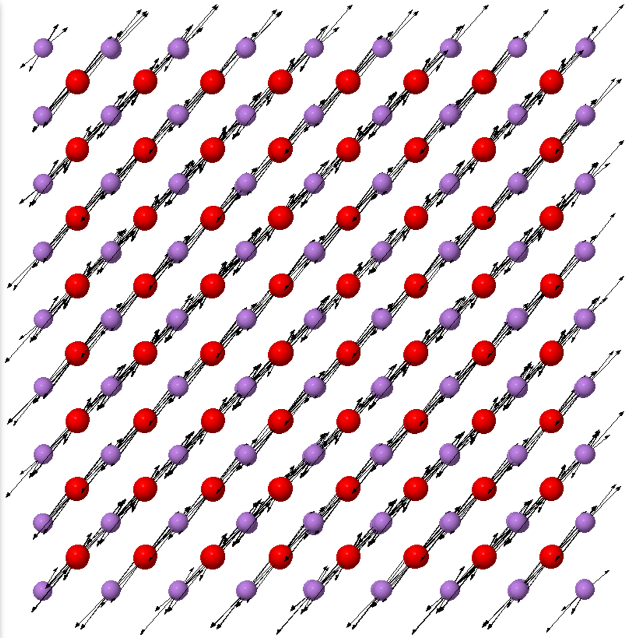
1-2% impurities causes 50% of propagating to be lost!



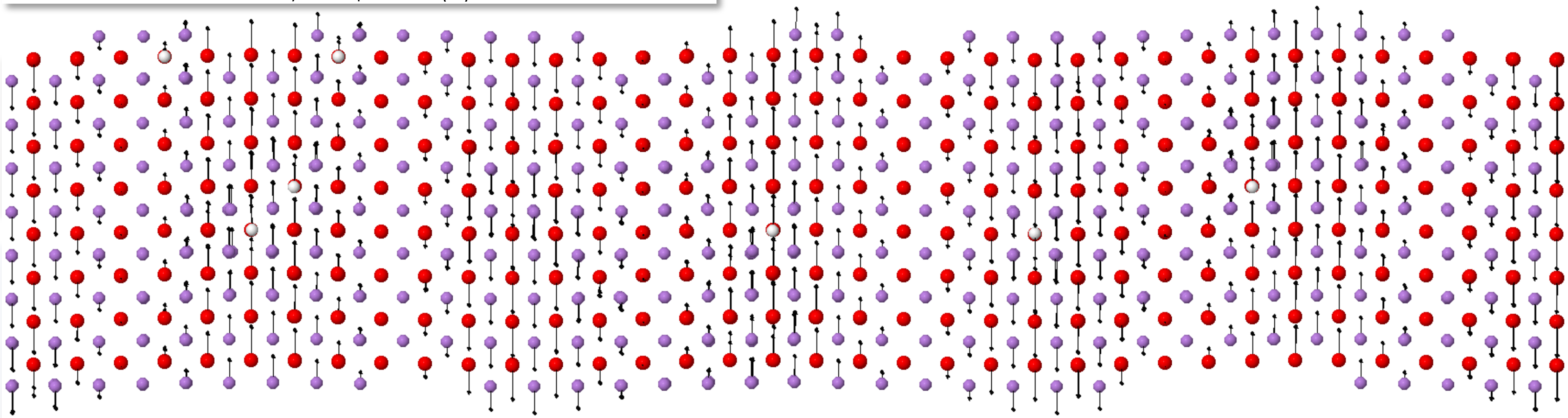


Evolution of Mode Character

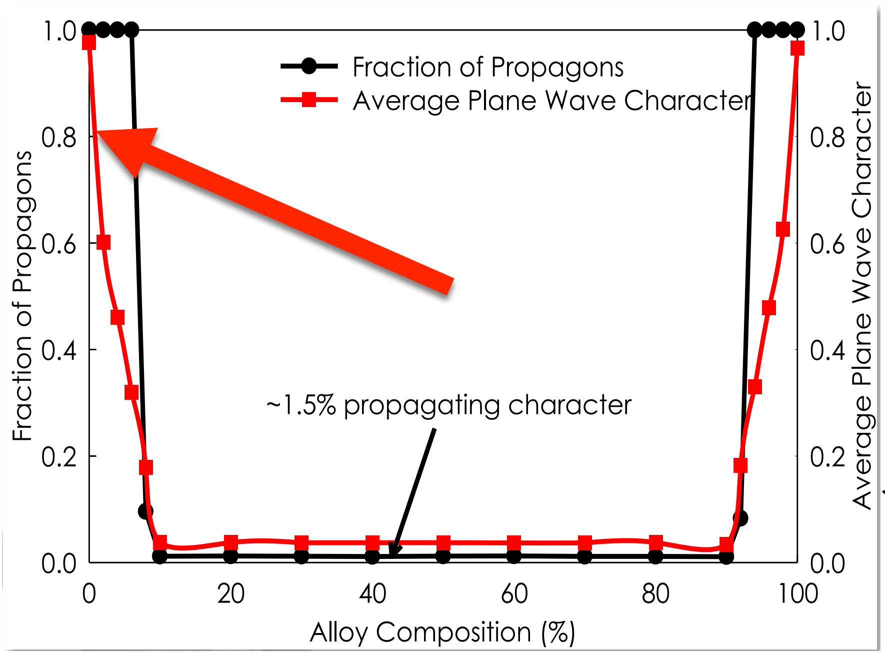


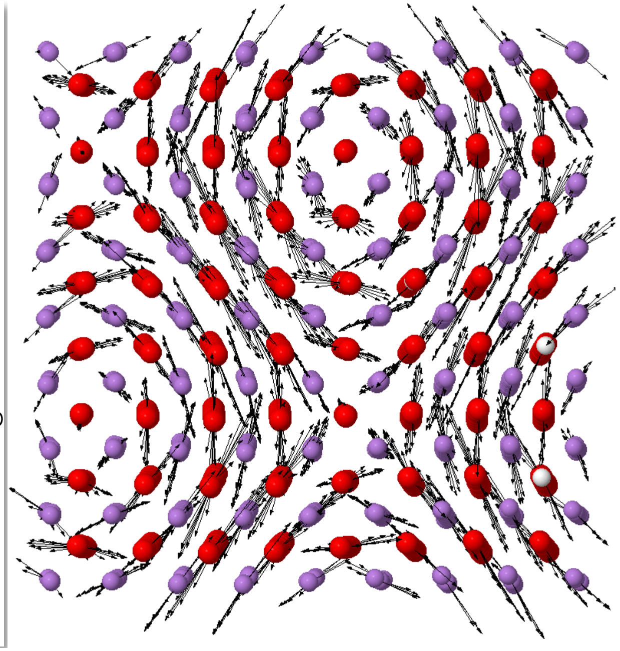


- 100%
- Periodicity
- Propagon

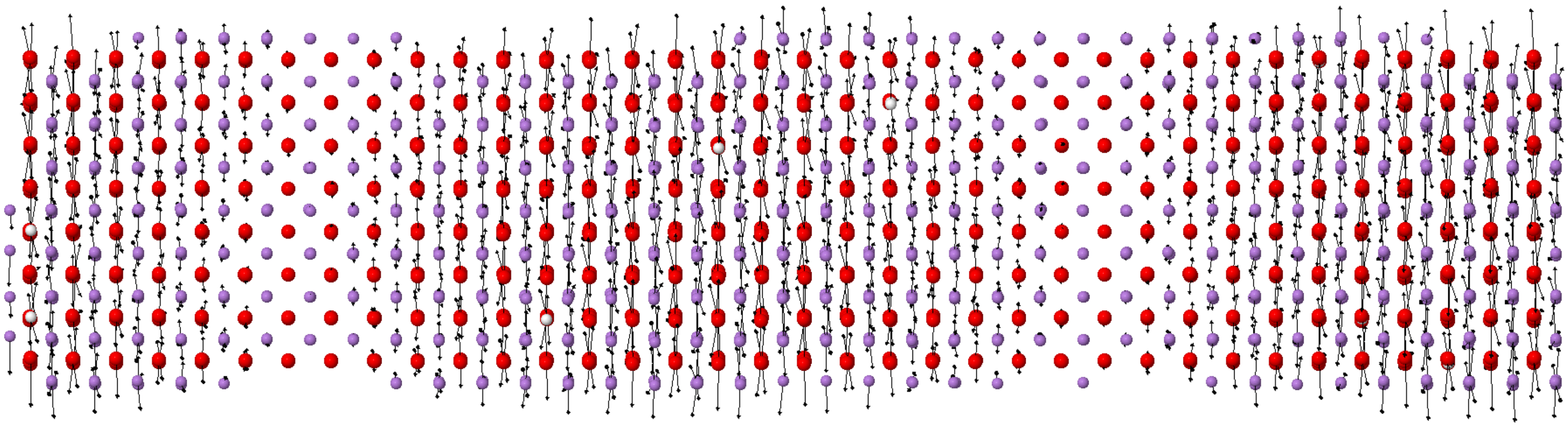


Evolution of Mode Character

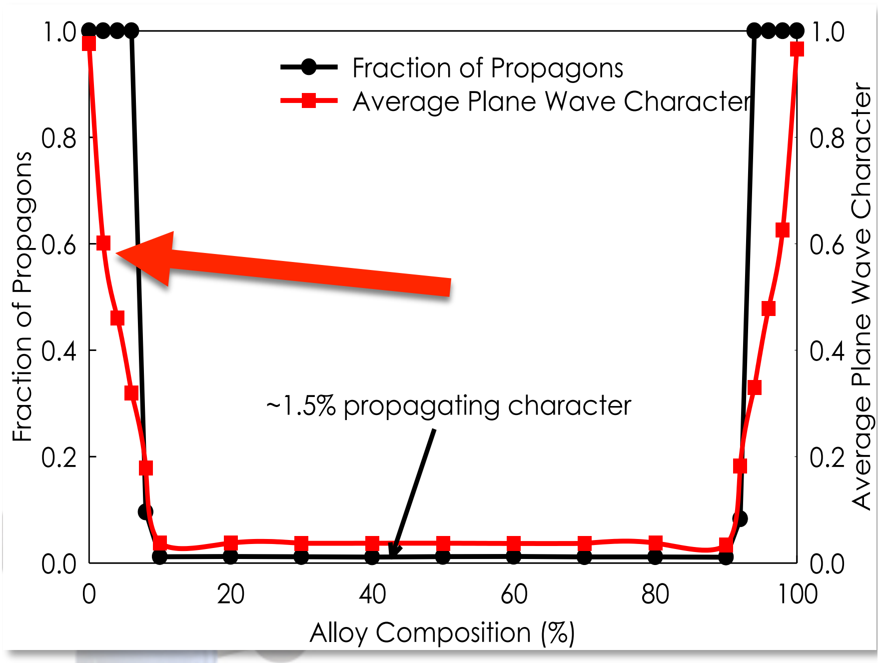


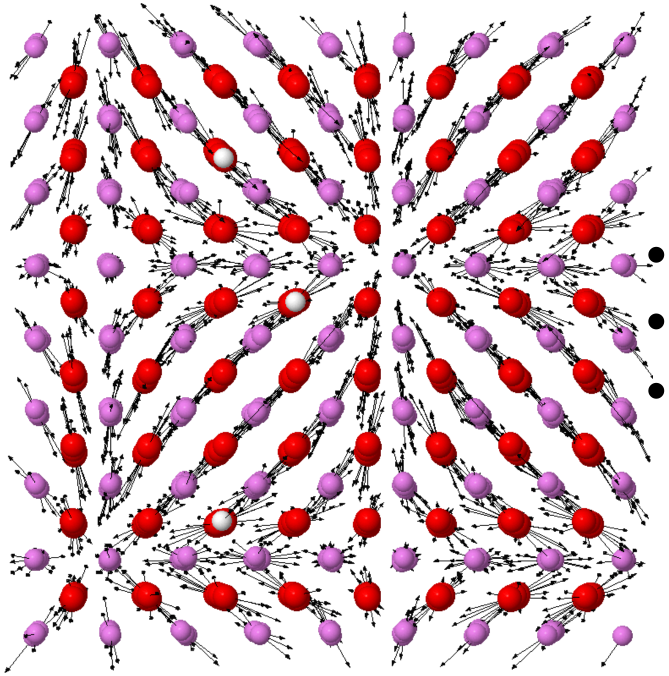


- 80%
- Periodicity
- Propagon

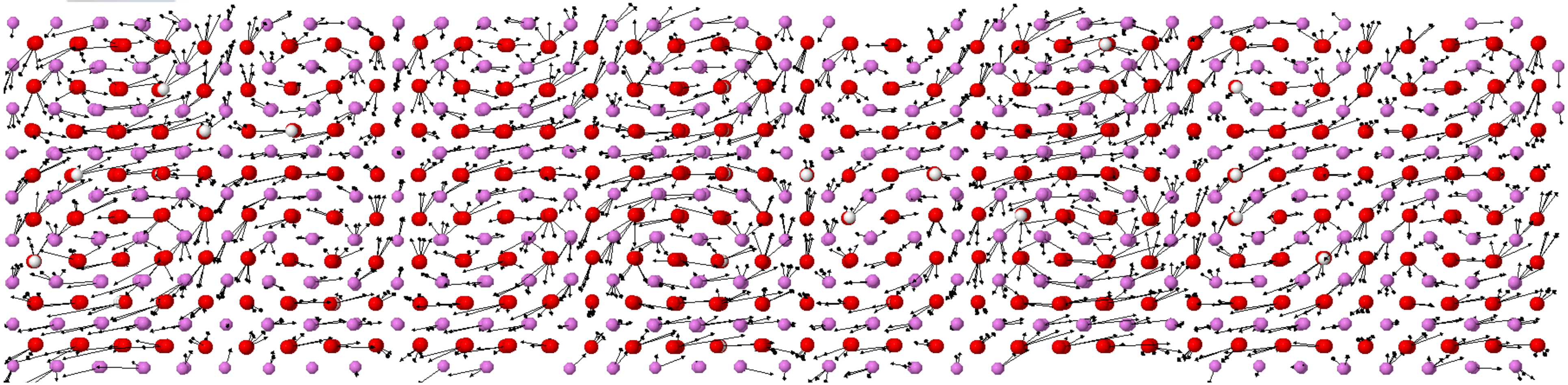


Evolution of Mode Character

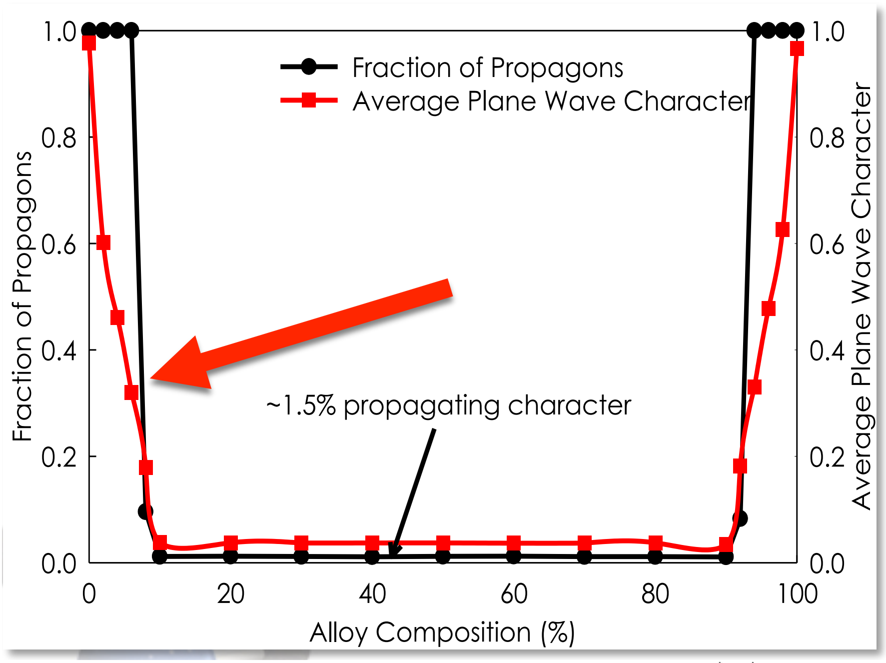


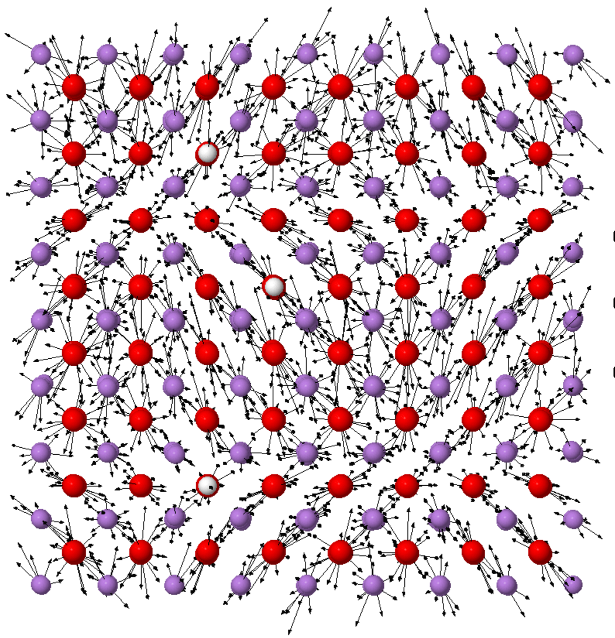


- 60%
- Periodicity
- Propagon

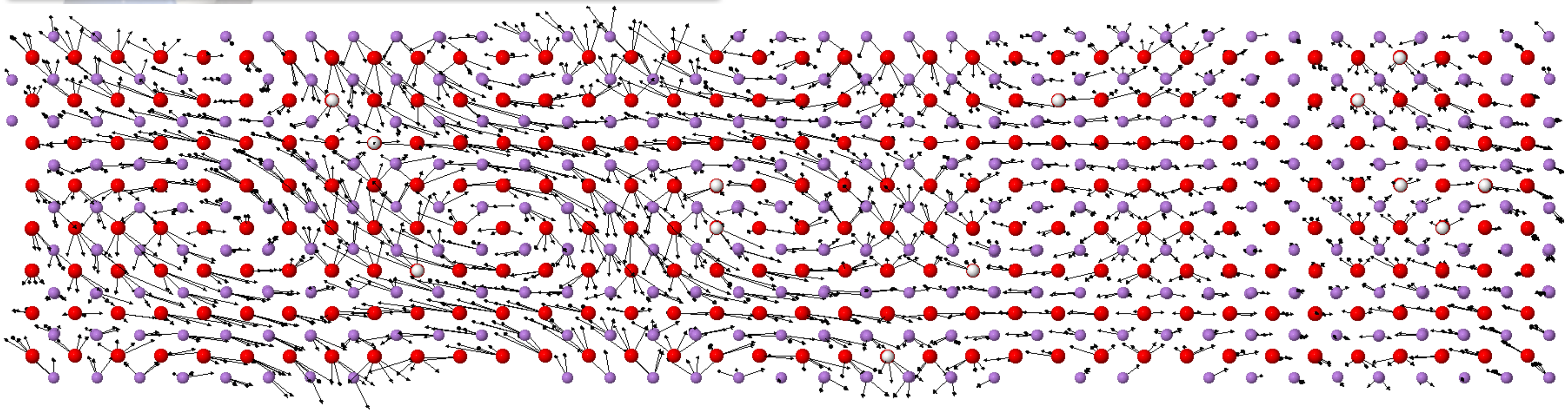


Evolution of Mode Character

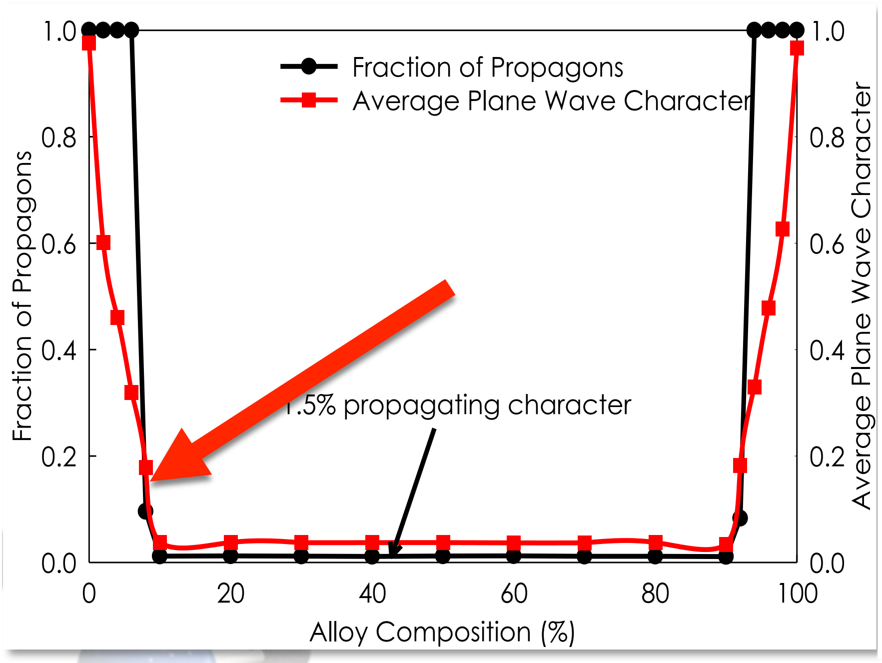


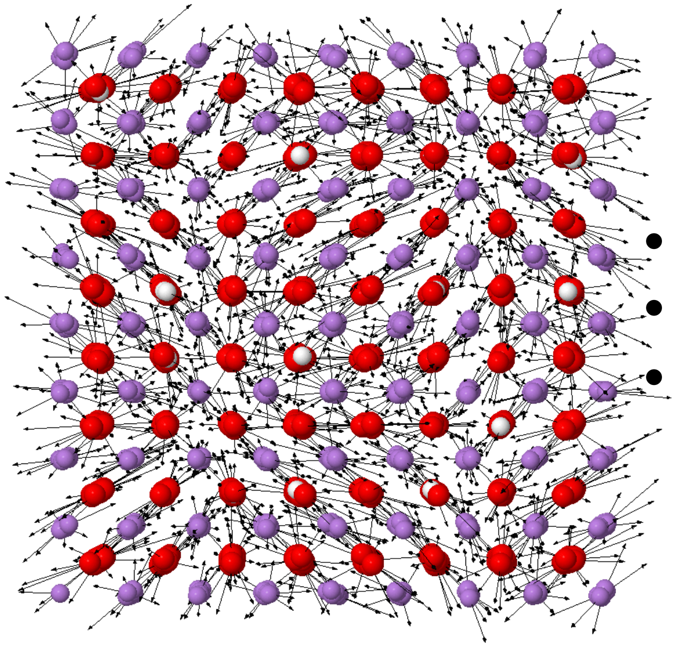


- 30%
- Periodicity
- Propagon

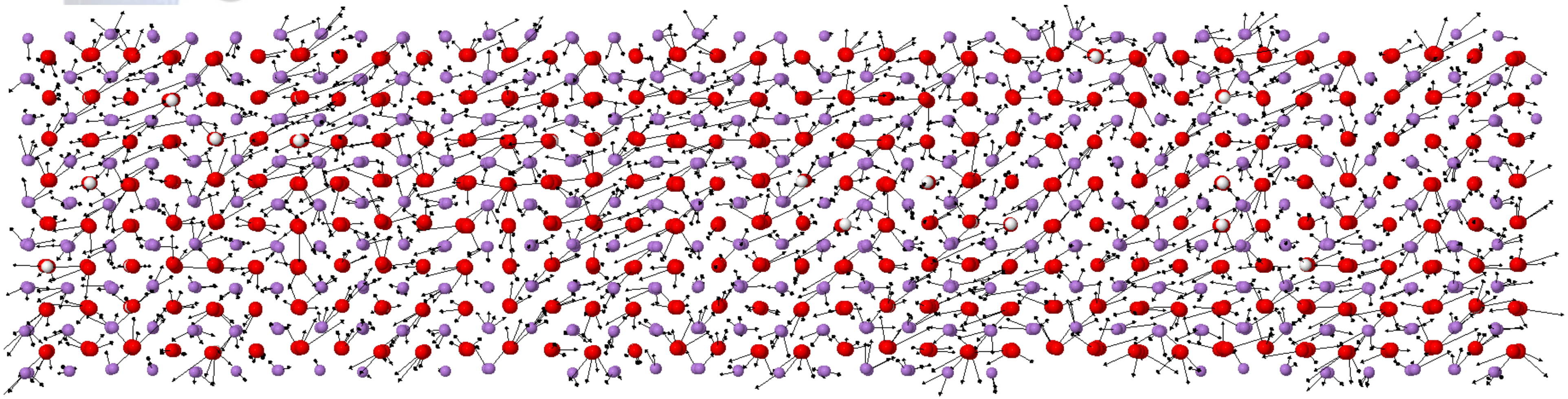


Evolution of Mode Character

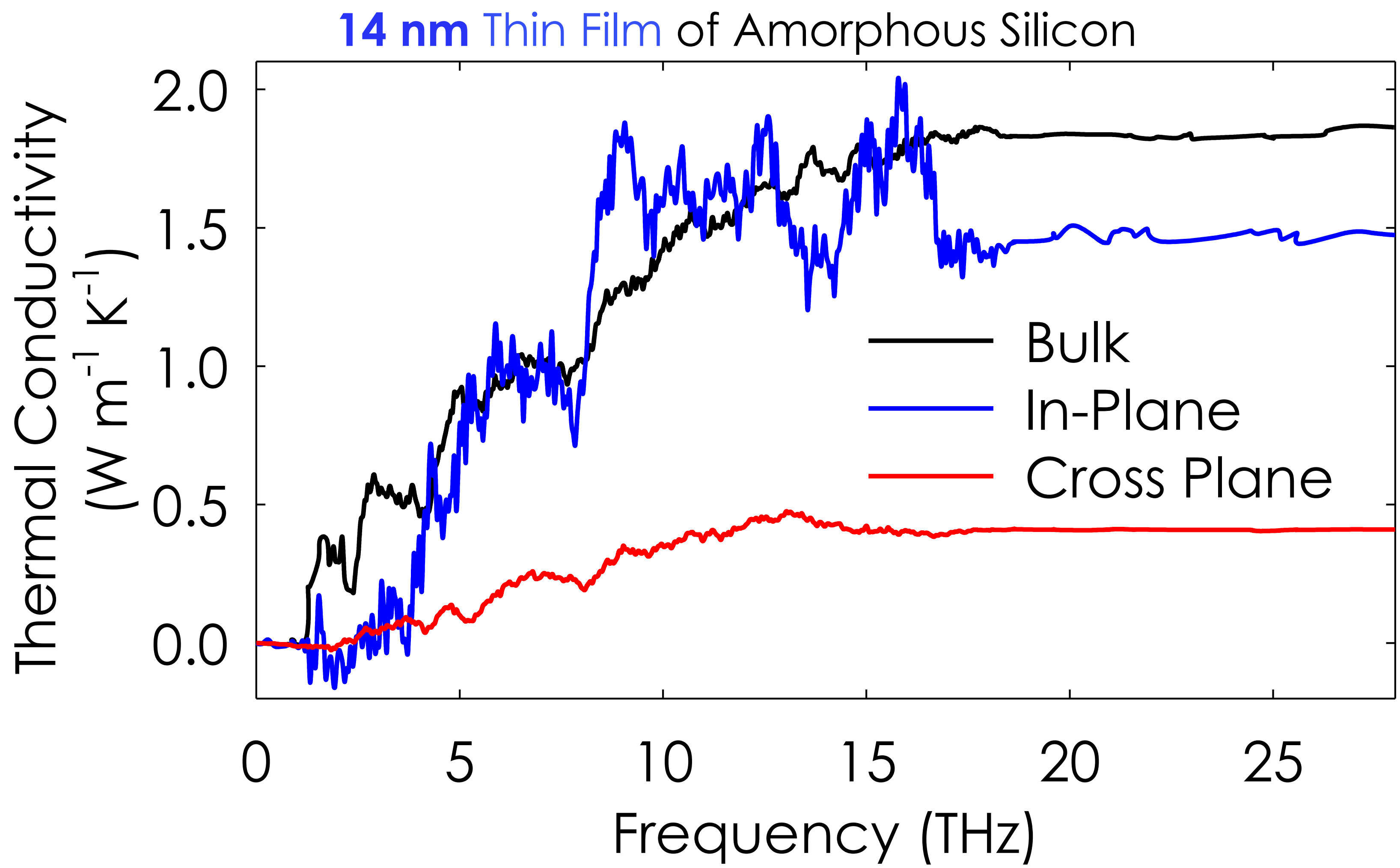




- 15%
- Periodicity
- Diffuson

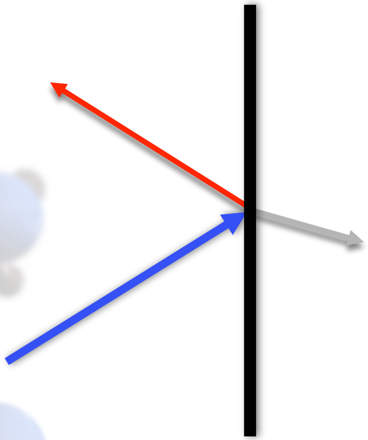


Diffuson Size Effects?



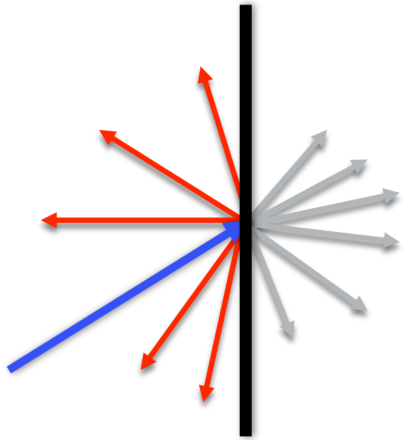
Interfaces: The Traditional View

Acoustic Mismatch Model



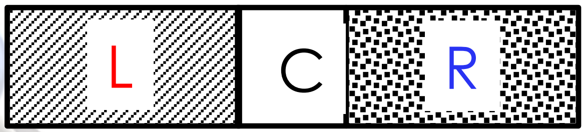
- Low temperatures
- Specular reflections
- No anharmonicity
- No atomic details

Diffuse Mismatch Model



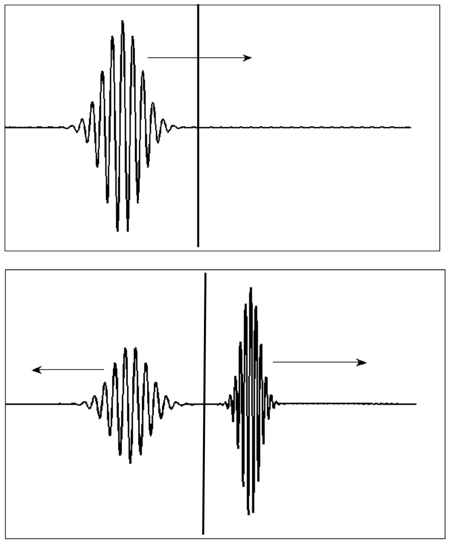
- Fully diffuse scattering
- High temperatures
- Partially anharmonic
- Partially atomic details

Atomistic Green's Function



- Low temperatures
- Partially anharmonic
- With atomic details

Wave Packet Method



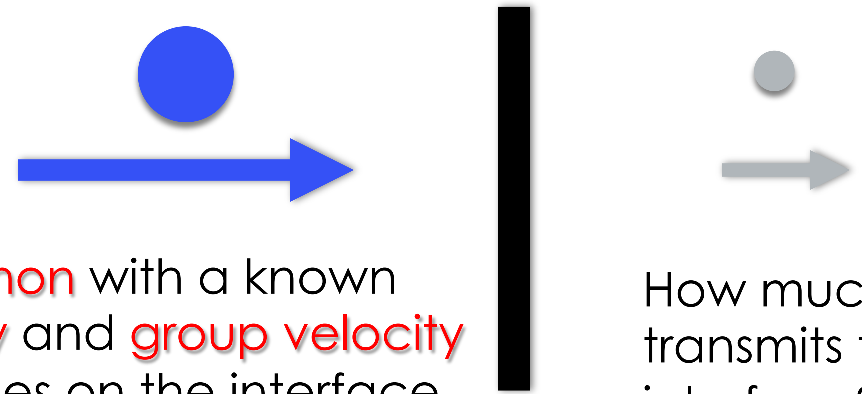
- Low temperatures
- Partially anharmonic
- With atomic details

PGM → Landauer Formalism

Transmission Probability

$$G = \frac{1}{4}\sum_j \int_0^{\omega_j^c} v_j(\omega)\, \alpha_j(\omega)\, \hbar\omega D_j(\omega) \frac{\partial n(\omega, T)}{\partial T} d\omega$$

Detailed balance → Only need modes from one side!



A phonon with a known energy and group velocity impinges on the interface

How much of its energy transmits through the interface?

There is an intrinsic maximum in transmission = 100%.

This means…

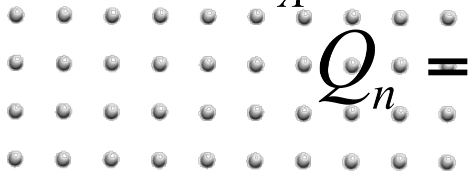
There is an intrinsic maximum in conductance →

Transmission = 100% for every mode

The Atomistic View – Which Modes?

{A}

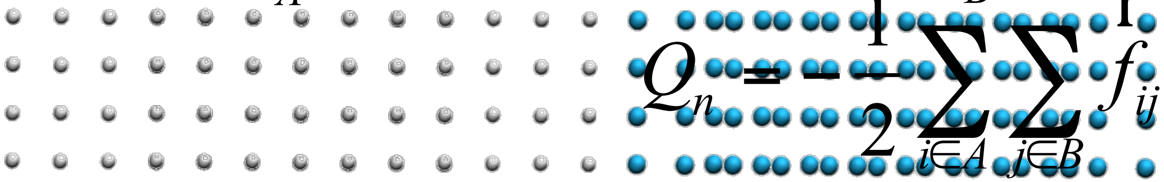
$3N_A$



$$Q_n = -\sum_{i\in A}\sum_{j\in B}\vec{f}_{ij}\cdot\vec{v}_{i,n}$$

{A+B}

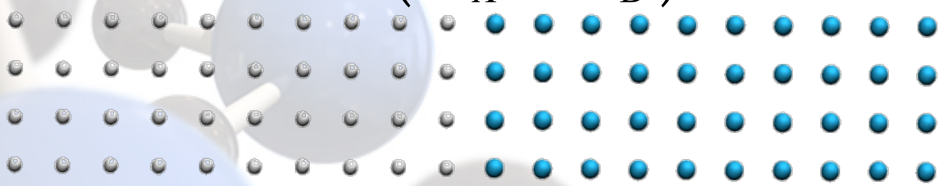
$3N_A$ $3N_A + 3N_B$ $3N_B$



$$Q_n = -\frac{1}{2}\sum_{i\in A}\sum_{j\in B}\vec{f}_{ij}\cdot\left(\vec{v}_{i,n}+\vec{v}_{j,n}\right)$$

{AB}

$3\left(N_A + N_B\right)$



$$Q_n = -\frac{1}{2}\sum_{i\in A}\sum_{j\in B}\vec{f}_{ij}\cdot\left(\vec{v}_{i,n}+\vec{v}_{j,n}\right)$$

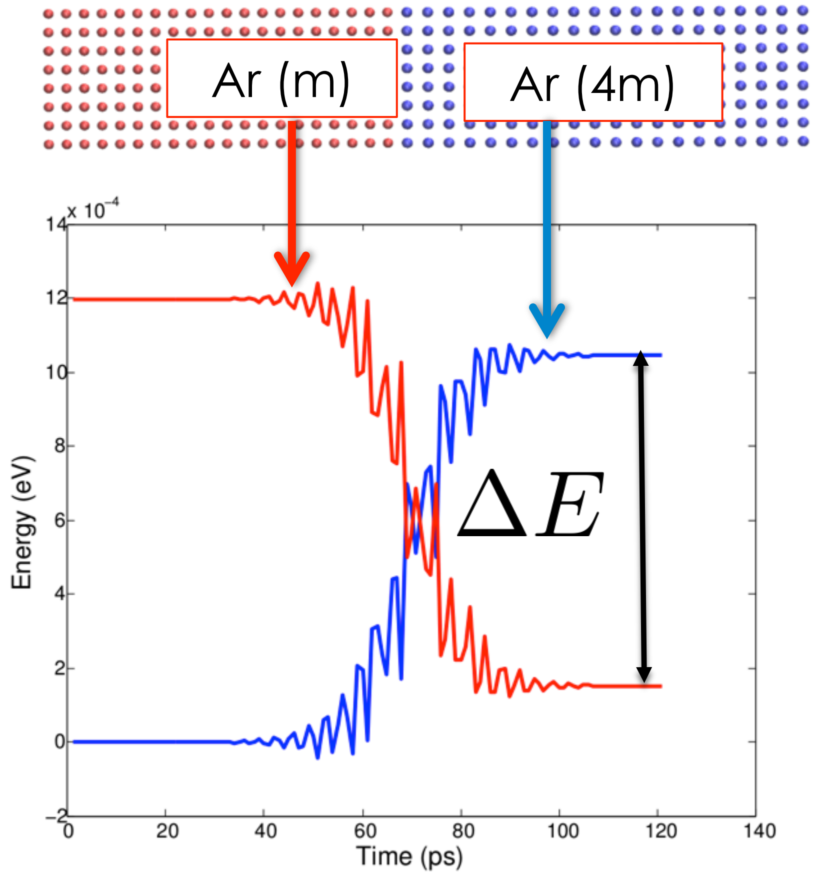
Which Coordinates are Right?

1. As the WP approaches $Q(t) = 0$
   $Q_n(t)$ must correspond to WP

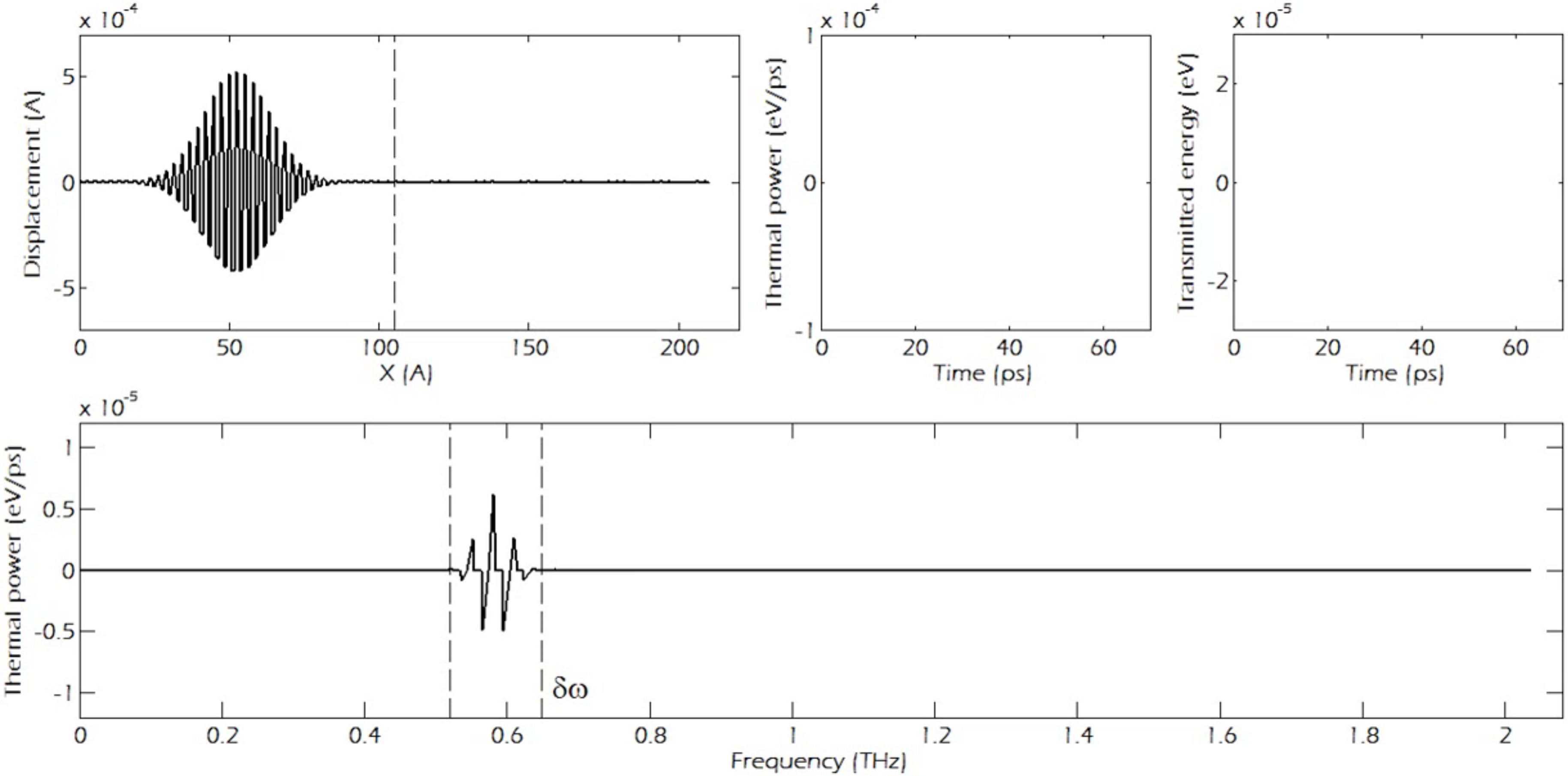
2. $\sum_n \int Q_n(t)dt = \Delta E$

3. No outside excitation

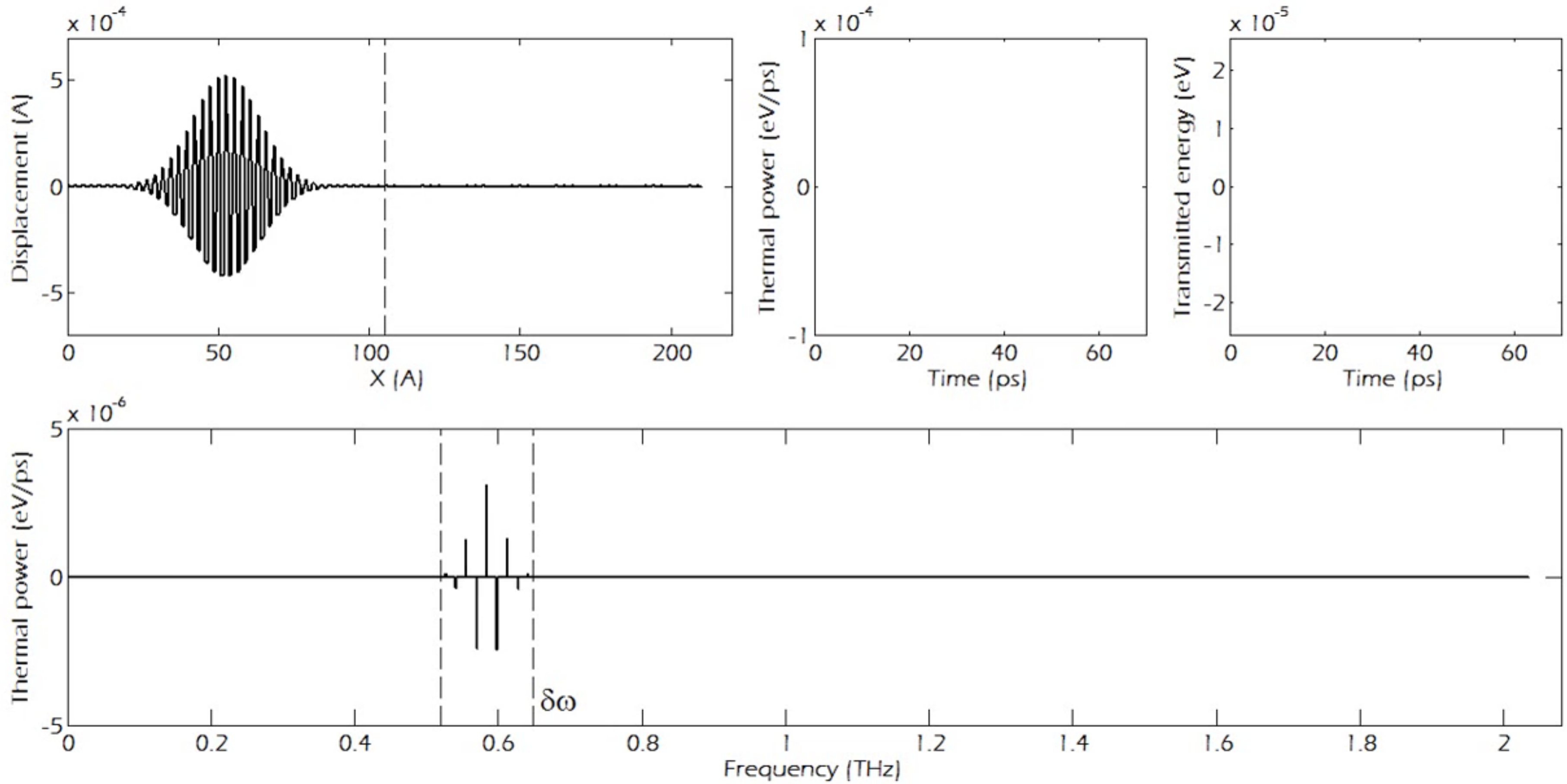
4. No outside contribution



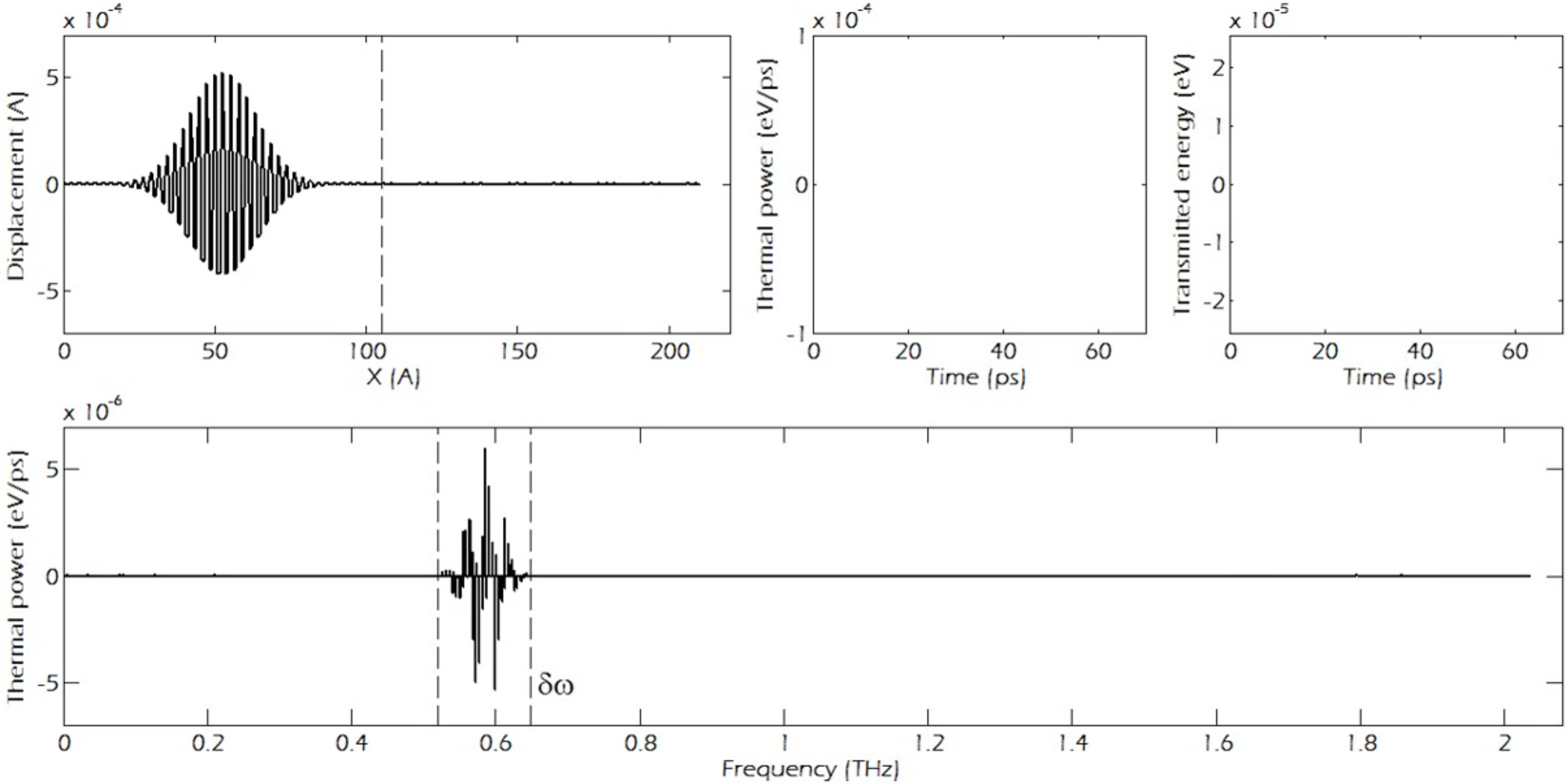
Wave Packet Test {A}



Wave Packet Test {A+B}



Wave Packet Test {AB}



Wave Packet Test