

Predicting Vibrational Mean Free Paths in Amorphous Materials

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Understanding thermal transport in crystalline systems requires detailed knowledge of phonons, which are the quanta of energy associated with atomic vibrations. By definition, phonons are non-localized vibrations that transport energy over distances much larger than the atomic spacing. For disordered materials (e.g., alloys, amorphous phases), with the exception of very long wavelength modes, the vibrational modes are localized and do not propagate like phonons. The Einstein model assumes that the mean free path of these localized vibrations is the average interatomic distance and that their group velocity is equal to the speed of sound. The Cahill-Pohl model assumes that the mean free path of the localized modes is equal to half of their wavelength. While these approach can be used to estimate the thermal conductivity of disordered systems, they only provide a qualitative description of the vibrations that contribute to the lattice thermal conductivity. Using lattice dynamics calculations and molecular dynamics simulations on model amorphous silicon and silica, we predict and characterize the contributions from phonons and localized vibrations to lattice thermal conductivity. The vibrational mean free paths are predicted for these two amorphous materials and the thermal conductivity accumulation function is compared with recent experimental results.

I. INTRODUCTION

The thermal conductivity of amorphous solids display unique temperature dependance compared to ordered solids.¹ Cahill argued that the lattice vibrations in a disordered crystal are essentially the same as those of an amorphous solid.²

Measurements by all the refs from Galli paper, including Moon.³⁻⁹ The key to understanding such measurement is to estimate a MFP for the vibrational modes in disordered systems.

The goal of this work is to predict the MFP of vibrational modes in disordered systems. Simple Lennard-Jones systems will be studied. A perfect LJ crystal are alloyed with a species of differing mass and amorphous samples are prepared. Thermal transport will be studied to quantify and characterize the ordered and disordered contributions to lattice thermal conductivity. In particular, a more rigorous way to classify vibrational modes in disordered alloys and amorphous samples as phonon-like or diffuson will be investigated. These results will be compared to the phenomenological Einstein and Cahill-Pohl models,^{???}.

The vibrational modes in these systems are characterized in the limit of propagating (phonon) and non-propagating (diffuson) modes by predicting the mode lifetimes and estimating their mean free paths. Estimating an effective dispersion relation is necessary for calculating an effective group velocity for disordered, which is crucial for transforming lifetimes to MFPs. The spectrum of phonon MFPs and the accumulated thermal conductivity

are predicted for a model of amorphous silicon. Predictions of thermal conductivity using a boundary scattering model demonstrates

II. THEORETICAL FORMULATION

A. Phonons

\mathbf{k} = sum over modes

For a perfect system, all vibrational modes are phonons.

$$k_{vib,\mathbf{n}} = \sum_{\mathbf{\kappa}} \sum_{\nu} c_{ph}(\mathbf{\kappa}_{\nu}) \mathbf{v}_{g,\mathbf{n}}^2(\mathbf{\kappa}_{\nu}) \tau(\mathbf{\kappa}_{\nu}). \quad (1)$$

Of particular interest is the phonon mean free path (MFP),

$$\Lambda(\mathbf{\kappa}) = |\mathbf{v}_{g,\mathbf{n}}^2(\mathbf{\kappa}_{\nu})| \tau(\mathbf{\kappa}_{\nu}), \quad (2)$$

which requires a group velocity.

Taking the phonon mode specific heat to be $c_{ph}(\mathbf{\kappa}_{\nu}) = k_B$, the phonon mode specific vibrational conductivity (Eq. (??)) can be written as

$$k_{vib,\mathbf{n}} = \sum_{\mathbf{\kappa}} \sum_{\nu} k_B D_{ph}(\mathbf{\kappa}_{\nu}), \quad (3)$$

and the vibrational conductivity is determined by the phonon mode diffusivities, defined as

$$D_{ph}(\kappa) = v(\kappa)^2 \tau(\kappa). \quad (4)$$

B. Diffusons

For disordered systems, the vibrational modes are no longer pure plane-waves (i.e., phonon modes), except in the low-frequency (long-wavelength) limit. When applied in the classical limit, the Allen-Feldman (AF) theory computes the contribution of diffusive, non-propagating modes (i.e., diffusons) to thermal conductivity from¹⁰

$$k_{AF} = \sum_{diffusons} \frac{k_B}{V} D_{AF,i}(\omega_i), \quad (5)$$

where $D_{AF,i}$ is the mode diffusivity and ω_i is the frequency of the i th diffuson. The diffusivity of diffusons can be calculated from harmonic lattice dynamics theory.¹⁰⁻¹²

The relative contribution of both phonons and diffusons to the total vibrational conductivity has been estimated to be approximately equal for a-Si,¹³ while earlier studies find that k_{ph} is substantially less.¹²

C. Thermal Diffusivity Limits

$k = \sum c D$, only for low- ω , long- λ limit does $D = 1/3 v^2 \tau$, or $1/3 v \lambda$

“The diffusivity D_i cannot be meaningfully represented as $v_i^2 \tau_i / 3$ since v_i and τ_i cannot be independently defined. Numerically we find that D_i for diffusons is of order $D a / 3$ where a is the interparticle spacing. Also, D_i is independent of the particular state i , depending only on the energy as seen in Fig. 12. Our Eq. 7 for the heat conductivity is a close analog of Eq. A4.”

$$\tau = \frac{2\pi}{\omega}. \quad (6)$$

$$\Lambda = \lambda. \quad (7)$$

It is not clear that these are equivalent limits if the group velocity in Eq is mode-dependent.

It was noticed by Birch and Clark (1940), and by Kittel (1948) that in glasses (T_g) at $T \sim 20K$ could be interpreted as the specific heat $C(T)/V$ multiplied by a temperature-independent diffusivity D of order $a^2 D / 3$ where a is an interatomic distance. In the phonon-gas model, this would correspond to $l \sim a$, too small to justify use of the model. The success of this observation implies that the dominant normal modes in a glass are of the D variety, not P because P implies $l \gg a$, and not L

because L implies $D = 0$ until anharmonic corrections are added which make D depend on T . This successful (and we believe, essentially correct) interpretation lost favor after Anderson localization was understood, because a misconception arose that the P/D boundary (which certainly lies low in the spectrum of a glass) should lie close to the E/L boundary.

Predictions for a-Si, also effects of mass disorder.¹¹: is D_i pinned near a value of $D_i \sim (1/3)va$?

$$D_{IR} = \frac{2\pi}{3} \frac{v_s^2}{\omega}. \quad (8)$$

$$D_{HS} = \frac{1}{3} v_s a, \quad (9)$$

Assuming that all vibrational modes travel with the sound speed, v_s , and scatter over a distance of the lattice constant, a , a high-scatter (HS) limit of thermal conductivity in the classical limit is¹⁴

$$k_{HS} = \frac{k_B}{V_b} b v_s a, \quad (10)$$

where V_b is the volume of the unit cell and b is the number of atoms in the unit cell. The HS limit will be used to discuss the differences between the LJ argon and SW silicon alloys.

III. CALCULATION DETAILS

A. Sample Preparation

1. Amorphous Si

We use models created by the WWW algorithm as described in⁷.

get samples for a-Si (from AF). Sample sizes of 216, 1000, 4096, and 100,000 atoms.

samples were originally created using WWW algo. The SW potential is used with these samples. With amorphous materials, have to deal with meta-stability. There are many potential energy configurations (atomic positions) which are nearly equivalent in energy. At a sufficient temperature, the meta-stable states cause the atomic positions to vary. This has an effect on the prediction of the vibrational mode lifetimes when using the normal mode decomposition method in the time domain.

Anneal at 1100 K for 5 ns to remove meta-stability. The meta-stability is demonstrated by increased v_s after annealing. structural relaxation: $U_l = 7.64 \times 10^{-4}$ m/s and $U_T = 3.67 \times 10^{-4}$, indicates that there has been structural relaxation $vs_{long} : 8.2715e + 03, vs_{tran} : 3.8867e + 03$

A large sample (800,000 atoms) was created from the 100,000 atom sample by treating it as a unit cell and tiling the 800,000 atom supercell. The box size is nm.

All calculations based on density of the perfect crystal with lattice constant 5.43 Ang.

2. Amorphous SiO_2

samples from Alan. The 24-6 LJ potential is replaced with a 12-6, which has a negligible effect on the predictions presented in this paper.

New samples of size up to 4400 atoms were created by first tiling the smaller samples, and then performing a liquid quench procedure similar to Alan. Results for these quenched, fully amorphous samples agree closely with those of the unquenched, tiled samples.

B. Simulation Details

The MD simulations were performed using LAMMPS.¹⁵

$dt = 0.00905$, $dt = 0.0005$

20 for 10 seeds. For the GK method, the thermal conductivity of SiO_2 and s-Si, a interval of the the HCACF integral can be found which is constant within the statistical noise.

The trajectories from the MD simulations used for the GK calculations (Setion) are also used in the normal mode decomposition method (Section).

The AF calculation is performed using the package GULP. A Lorentzian broadening is used of $5\delta\omega_{avg}$ and $14\delta\omega_{avg}$ for a-Si and a-SiO₂. Varying the broadening around these values does not change the resulting thermal conductivity significantly (see Section).

IV. VIBRATIONAL PROPERTIES

A. Density of States

Feldman measure the DOS using the average level spacing.¹²

$$DOS(\omega) = \sum_i \delta\omega_i - \omega, \quad (11)$$

where a unit step function is used to broaden $\delta(\omega_i - \omega)$. Debye Model:

$$DOS(\omega) = \frac{3 * \pi \omega^2}{2v_s^3}, \quad (12)$$

See it clearly for a-Si, only barely for a-SiO₂ estimates of the sound speeds from the elastic constants are

A fit of the DOS gives

$$v_s = \frac{1}{3}v_{s,L} + \frac{2}{3}v_{s,T}, \quad (13)$$

$$v_s = \frac{B_{mod}}{\rho}, \quad (14)$$

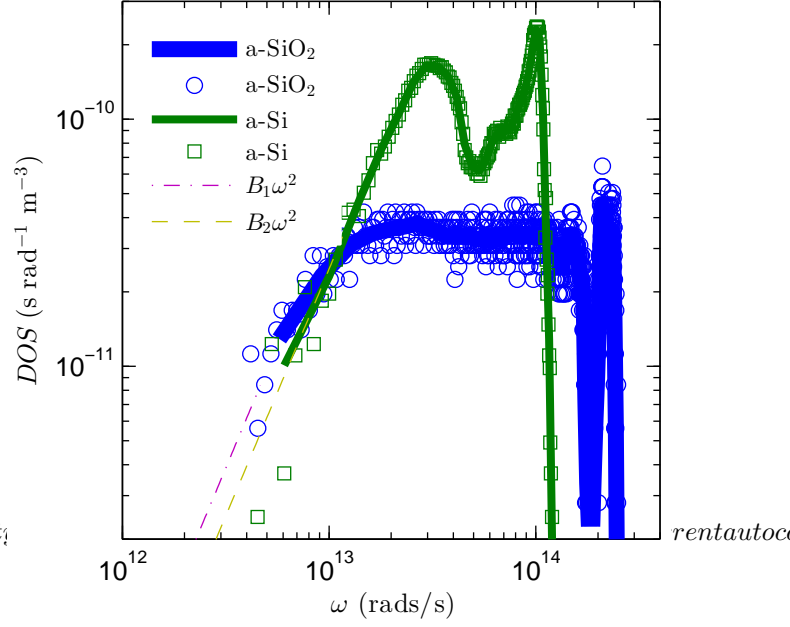


FIG. 1: film thickness dependant thermal conductivity of a-Si from experiment.

“The vertical lines starting on the frequency axis give the inverse participation ratios of the states, showing the quasilocized or resonant nature of the low- est eigen-state at 4.2 meV.”

“The modes of mixed character which lie outside or in between the central groups of pure plane-wave modes are typically resonances as indicated by the large values of $1/p_i$. However, these modes are also reasonably well interpreted as filling in the appropriate tails of Lorentzian response functions, modulo small statistical fluctuations to be expected in finite systems.

bigger and small Q s become less sparse, Lorentzians of fixed width will overlap increasingly, and can force out the resonant states which otherwise would inhabit the gaps. For Q of order $1/L$ there will always be gaps, no matter how big the system L see for instance the region near 4.1 meV in Fig. 6 where a resonance occurs, but these gaps drift toward $Q \rightarrow 0$ and 0 as L increases. Therefore the distinction between special frequencies lying in gaps, and other frequencies lying in Lorentzian peaks, must disappear as L increases. There are two possibilities: either resonant behavior entirely disappears, or else it remains in a diluted form and is shared uniformly by all the normal modes. That is, at any given frequency there may be isolated parts of a large sample which are particularly sensitive to oscillation at just this frequency and temporarily trap selected traveling waves of this frequency. If this behavior is found for all normal modes, then any one normal mode will be freely propagating almost everywhere, and it becomes a subtle matter of definition or taste whether they should

be called resonances at all. ”

B. Structure Factor

Fig 4 of this work shows a dispersion extracted by locating the peaks in the structure factor.¹⁶

Fig. 5 discusses how since the low freq modes are sparse, there is a resonant effect between¹²

If all modes are summed over, this gives the frequency spectrum needed to construct a (nonstationary) propagating state with a pure wave vector \mathbf{Q} and pure longitudinal or transverse polarization¹¹. Locations of spectral peaks are peaked like a acoustic dispersion branches. Only low-frequency vibrations have an (approximate) wavevector in disordered systems, and there is no theorem guaranteeing this.¹²

However, it is very difficult to distinguish between localized and extended modes at high frequencies on the basis of their $S(k, \nu)$ functions, as illustrated by the very similar scattering functions for a 67-meV localized and a 63-meV extended mode in Fig. 3(b).¹⁷

The dynamic structure factor can be useful for demonstrating the plane-wave character of low-frequency vibrations. However, on a mode-by-mode basis, it is unable in general to characterize a given mode as either localized or delocalized. Thus, it is not possible to

$$\mathbf{v}_{g,n}(\boldsymbol{\kappa}) = \frac{\partial \omega(\boldsymbol{\kappa})}{\partial \boldsymbol{\kappa}}. \quad (15)$$

estimates of the sound speeds are found from using finite difference of the peaks in $S_{T,L}$ for different values of k .

The structure factor at a VC wave vector $\boldsymbol{\kappa}_{VC}$ is defined as¹⁸

$$S^{L,T} = \sum_{\nu} E^{L,T} \delta[\omega - \omega_{\nu}], \quad (16)$$

where the summation is over the Gamma modes, E^T refers to the transverse polarization and is defined as

$$E^L = \left| \sum_b \hat{\boldsymbol{\kappa}}_{VC} \cdot \mathbf{e} \exp[i\boldsymbol{\kappa}_{VC} \cdot \mathbf{r}_0^{(l=0)}] \right|^2 \quad (17)$$

and E^L refers to the longitudinal polarization and is defined as

$$E^T = \left| \sum_b \hat{\boldsymbol{\kappa}}_{VC} \times \mathbf{e} \exp[i\boldsymbol{\kappa}_{VC} \cdot \mathbf{r}_0^{(l=0)}] \right|^2. \quad (18)$$

In Eqs. (17) and (18), the b summations are over the atoms in the disordered supercell, $\mathbf{r}_0^{(l=0)}$ refers to the equilibrium atomic position of atom b in the supercell, l labels the unit cells ($l = 0$ for the supercell), α labels the Cartesian coordinates, and $\hat{\boldsymbol{\kappa}}_{VC}$ is a unit vector. Explicit

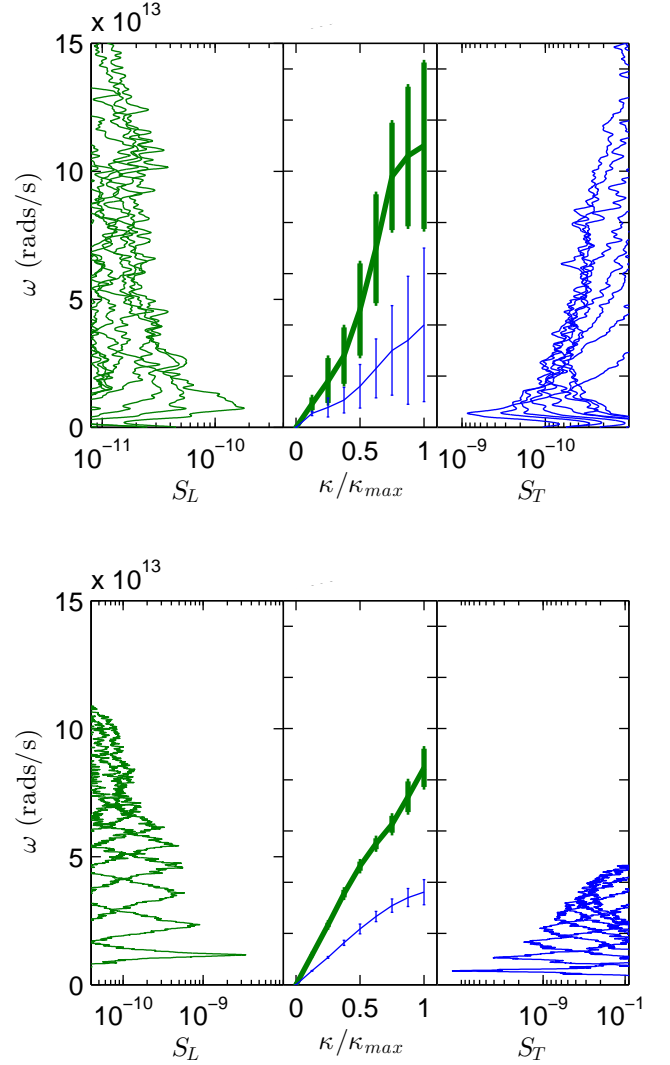


FIG. 2: film thickness dependant thermal conductivity of a-Si from experiment.

disorder is included in the Gamma frequencies ω and the $3N_a$ components of the eigenvectors, \mathbf{e} .

$$\mathbf{v}_{g,n}(\boldsymbol{\kappa}) = \frac{\partial \omega(\boldsymbol{\kappa})}{\partial \boldsymbol{\kappa}}. \quad (19)$$

A group velocity can be assigned by taking a slope of a graph of peak frequency versus wave vector. This velocity ($\approx 7 \times 10^3$ cm/sec) is slightly smaller than the acoustic velocity seen at smaller \mathbf{Q} 's. The mean free path is about the same as for the first peak, namely, the cell size. At larger wave vectors there is a dramatic change in the spectral shape. Insofar as they can be defined, full widths at half maximum are 30% of the peak frequency, which corresponds to mean free paths up to (with a generous choice of acoustic velocities) a factor of 2 or more smaller than

TABLE I: Estimated from the elastic constants, the pre-annealed group velocities are $v_{s,T,elas} = 3,670$, $v_{s,L,elas} = 7,840$, $v_{s,T,elas} = 2,541$, $v_{s,L,elas} = 4,761$

method	B_{mod} (Eq. (14))	$S_{T,L}$ (Eq. (16), (19))	DOS (Eq. (12))
a-SiO ₂			
transverse	3,161	2,732	2,339
longitudinal	5,100	4,779	
a-Si			
transverse	3,886	3,699	3,615
longitudinal	8,271	8,047	

the corresponding crystalline ~ 1 wavelengths. Clearly the mod spectrum are marginally propagating, whereas the major- of energies are wave vectors, are not propagating in any proper sense. nevertheless, they are extended contribute a significant heat current.¹¹

Table IV B.

C. Mode Lifetimes

Once the group velocities are predicted using the VC dispersion, the mode lifetimes are required to predict the thermal conductivity using Eq. (??). As an alternative to the VC-ALD approach for predicting lifetimes, which is discussed in the next section, we first use the MD simulation-based NMD method.^{19–22} In NMD, the atomic trajectories are first mapped onto the vibrational mode coordinate $q(\kappa; t)$ and its time derivative $\dot{q}(\kappa; t)$ by²³

$$q(\kappa; t) = \sum_{\alpha, b, l}^{3, n, N} \sqrt{\frac{m_b}{N}} u_{\alpha}(l; t) e^{*}(\kappa \frac{b}{\alpha}) \exp[i\kappa \cdot \mathbf{r}_0(l)] \quad (20)$$

and

$$\dot{q}(\kappa; t) = \sum_{\alpha, b, l}^{3, n, N} \sqrt{\frac{m_b}{N}} \dot{u}_{\alpha}(l; t) e^{*}(\kappa \frac{b}{\alpha}) \exp[i\kappa \cdot \mathbf{r}_0(l)]. \quad (21)$$

Here, m_b is the mass of the b_{th} atom in the unit cell, u_{α} is the α -component of the atomic displacement from equilibrium, \dot{u}_{α} is the α -component of the atomic velocity, and t is time. The total energy of each vibrational mode, $E(\kappa; t)$, is calculated from

$$E(\kappa; t) = \frac{\omega(\kappa)^2}{2} q(\kappa; t)^* q(\kappa; t) + \frac{1}{2} \dot{q}(\kappa; t)^* \dot{q}(\kappa; t). \quad (22)$$

The vibrational mode lifetime is predicted using

$$\tau(\kappa) = \int_0^{t^*} \frac{\langle E(\kappa; t) E \rangle}{\langle EE \rangle} dt, \quad (23)$$

Lifetimes in amorphous silicon predicted before using a normal mode approach, but mode-by-mode properties were not presented.²⁴

Lifetimes were predicted using anharmonic lattice dynamics, but no thermal transport properties were predicted.²⁵

Thermal diffusivity was predicted for a percolation network which showed Rayleigh type scattering dependence in the low-frequency limit.²⁶

Thermal diffusivity has been predicted using a wave-packet method

The lifetimes of vibrational modes in a-Si were predicted using normal mode decomposition.¹³

“To implement Eq. 6 one must know the correct Q dependence of κ . As shown in Fig. 8, the fitted values scatter too much to guide the extrapolation well. In principle, at very small Q one should get a form $\kappa \propto Q^4$ which corresponds to Rayleigh scattering of sound waves from the structural disorder. The data of Fig. 8 do not fit a Q^4 law; the Q^2 curve shown in the figure is a better fit. Two experiments^{13,15} but not a third³⁵ and one calculation¹⁸ on a-SiO₂ have also given Q^2 . We do not know a theory which can give this law in a harmonic model.

“The conclusion is that intrinsic harmonic glassy disorder contained in our finite calculation kills off the heat-carrying ability of propagons rapidly enough without invoking any exotic mechanism. Our $\kappa(T)$ curve is reminiscent of the experiments of Zaitlin and Anderson after holes are introduced to enhance the elastic damping of long-wavelength modes. The plateau disappears from their data in much the same way that it disappears from our theory due to extra damping of small- Q propagons.

D. Diffusivities

E. Mean Free Paths

V. THERMAL CONDUCTIVITY

A. Bulk

B. Accumulation

“We find that we cannot define a wave vector for the majority of the states, but the intrinsic harmonic diffusivity is still well-defined and has a numerical value similar to what one gets by using the Boltzmann result, replacing v by a sound velocity and replacing l by an interatomic distance a . ”¹¹

“In order to fit the experimental $\kappa(T)$ it is necessary to add a Debye-like continuation from 10 meV down to 0 meV. The harmonic diffusivity becomes a Rayleigh law and gives a divergent $\kappa(T)$ as $T \rightarrow 0$. To eliminate this we make the standard assumption of resonant-plus-relaxational absorption from two-level systems (this is an anharmonic effect which would lie outside our model even if it did contain two-level systems implicitly). ”¹¹

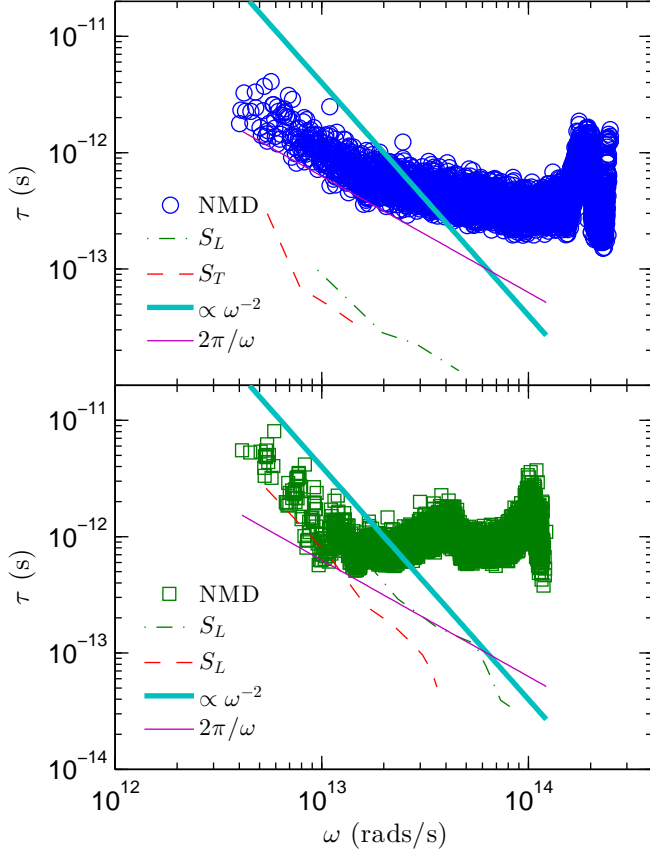


FIG. 3: film thickness dependant thermal conductivity of a-Si from experiment.

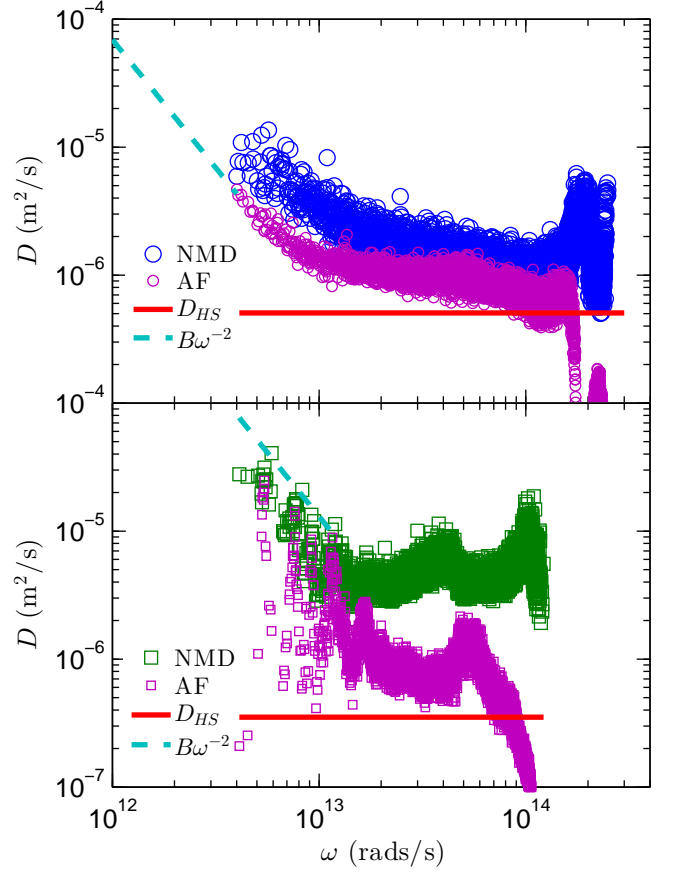


FIG. 4: film thickness dependant thermal conductivity of a-Si from experiment.

$$k = \int_0^{\omega_{max}} \frac{d\omega DOS(\omega) C(\omega/T) D(\omega)}{V} \quad (24)$$

$$C(\omega/T) = k_B \frac{(\hbar\omega/2k_B T)^2}{\sinh(\hbar\omega/2k_B T)} \quad (25)$$

Assuming the thermal conductivity form Eq. for the lowest frequency modes in the system, the thermal conductivity as a function of the system size takes the form

$$\frac{k(N_0)}{k_{bulk}} = 1 - \frac{c_0}{N_0}, \quad (26)$$

Debye model. $k_{tot} = k_{phonon} + k_{AF}$. For lack of a rigorous definition of phonon vs diffusion, we will define $k_{phon} = k_{debye}$.

C. Experimental Results

Table IV B.

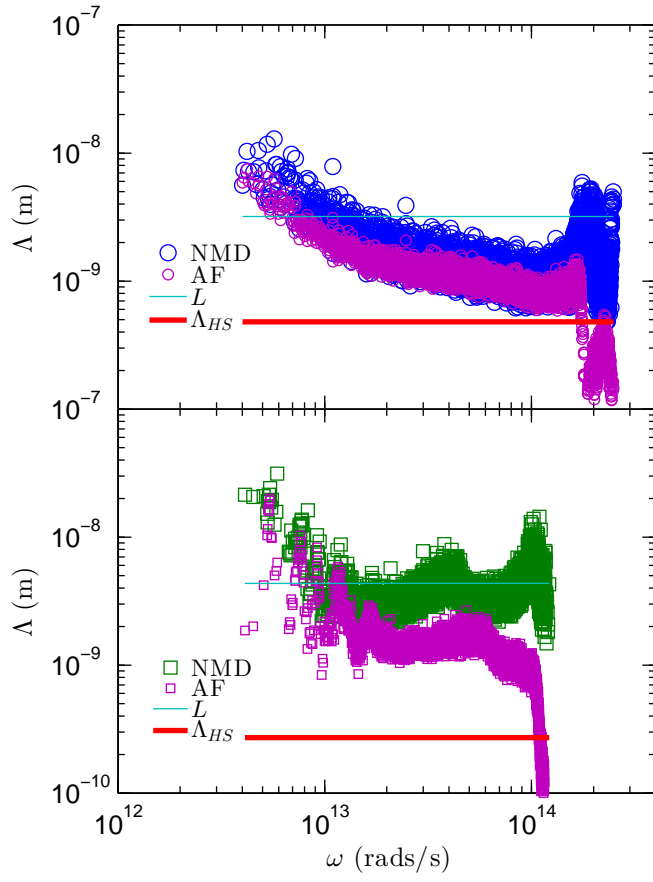


FIG. 5: film thickness dependant thermal conductivity of a-Si from experiment.

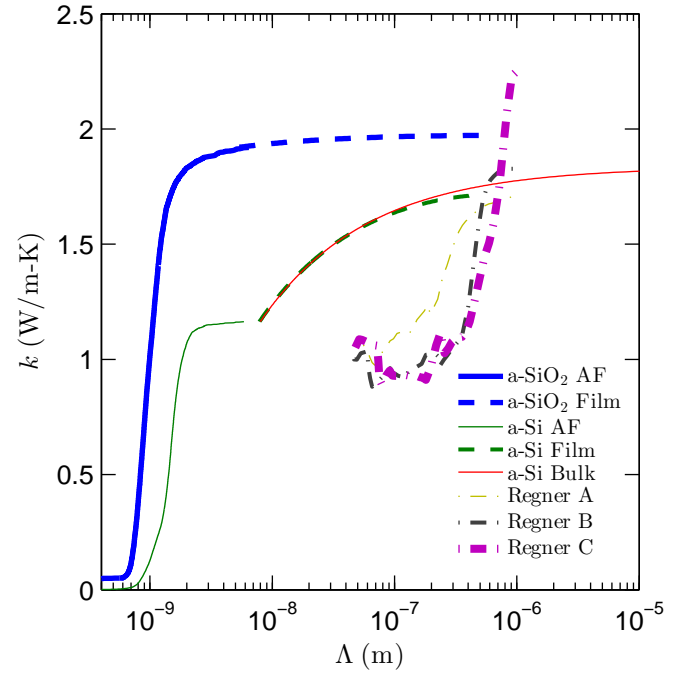
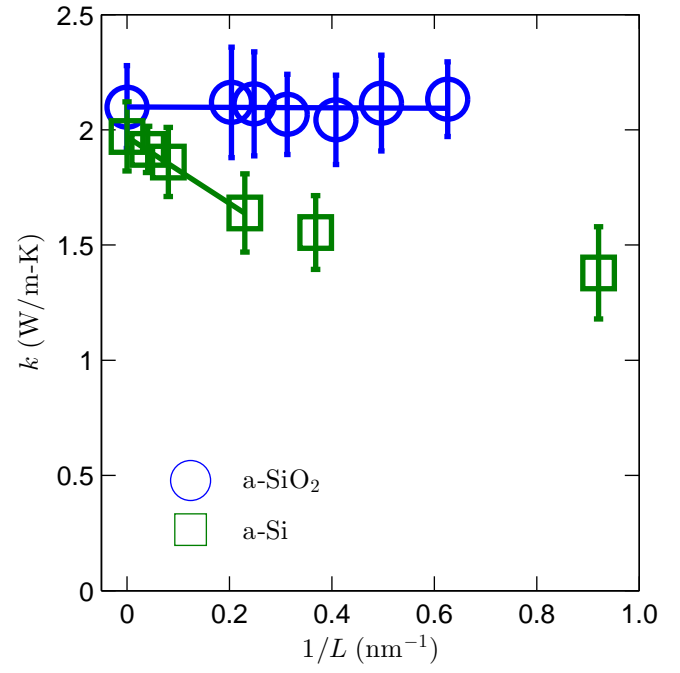


FIG. 7: film thickness dependant thermal conductivity of a-Si from experiment.

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