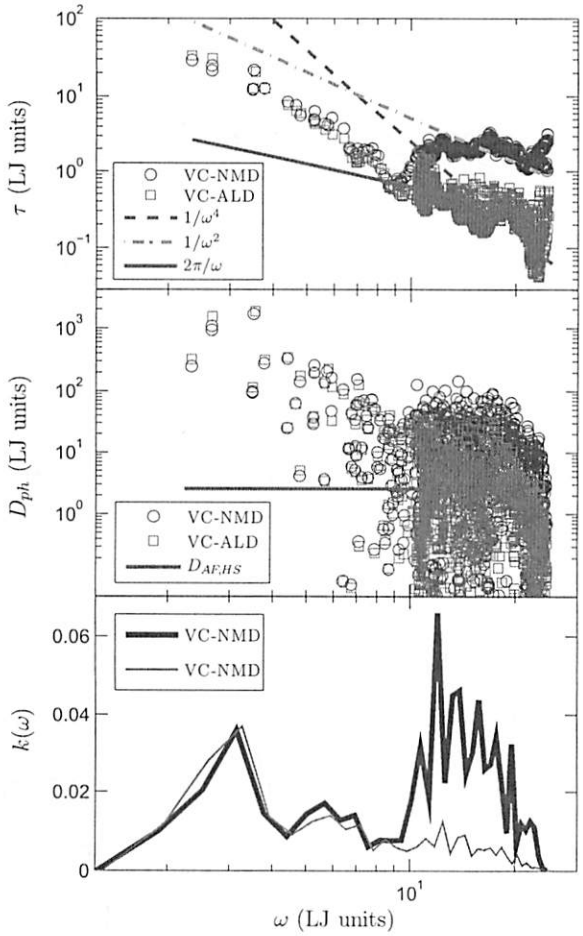


FIG. 5: (a) predicted lifetimes for VC modes using VC-NMD and VC-AlD for LJ argon. (b) predicted VC mode thermal diffusivities, compared to the AF,HS limit. (c) the thermal conducitivty frequency spectrum, which is peaked at high frequency, in contrast to SW silicon (Fig).

IV. THERMAL CONDUCTIVITY PREDICTIONS

The thermal conductivity can be now predicted using the vibrational mode properties predicted by the VC-NMD and VC-ALD methods. Given the discussion of the preceeding section, it is necessary to implement a third method for predicting thermal condudctivity. We choose the equilibirum MD-based green-kubo (GK) method.(cite) This method does not predict any mode-specific properties, and is thus a system-level prediction. Thermal conductivity predicted by GK has been shown to capture the effects of whatever scattering meachanisms are present in the MD simulation without any assumptions ~~(other than those that come~~

vague- be specific

by definition?

- get the point across that GK is "correct"

get all figs in section III, even if they get their own page

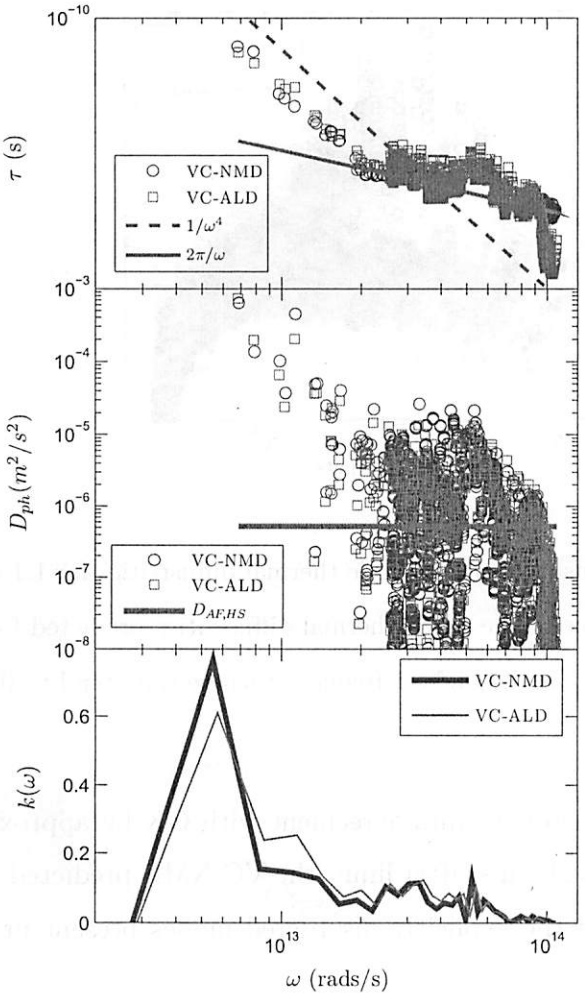


FIG. 6: (a) predicted lifetimes for VC modes using VC-NMD and VC-AlD for SW silicon. (b) predicted VC mode thermal diffusivities, compared to the AF,HS limit. (c) the thermal conducitivty frequency spectrum, which is peaked at low frequency, in contrast to LJ argon (Fig. 5).

the most 2 important point is how you specify the converged value of the integral

~~with the classical nature of the MD siulation).(cite)~~ Details of the GK and MD simulations are given in Appendix . For LJ argon, bulk thermal conductivity predictions are made for VC-NMD, VC-ALD, and GK (Fig. 8). For SW silicon, bulk thermal conductivity predictions can only be made for VC-ALD and GK (Fig. 9) because of the limited system size used for VC-NMD (see Appendix).

an earlier 2, section

For LJ argon, VC-NMD and VC-ALD underpredict the thermal conductivity compared to GK. The underprediction is only modest for VC-NMD, on the order of 20% or less for all

concentrations &. By adjusting the mode diffusivity as suggested in Section III D, the thermal conductivtiy

Introduce/motivate this p here instead of earlier

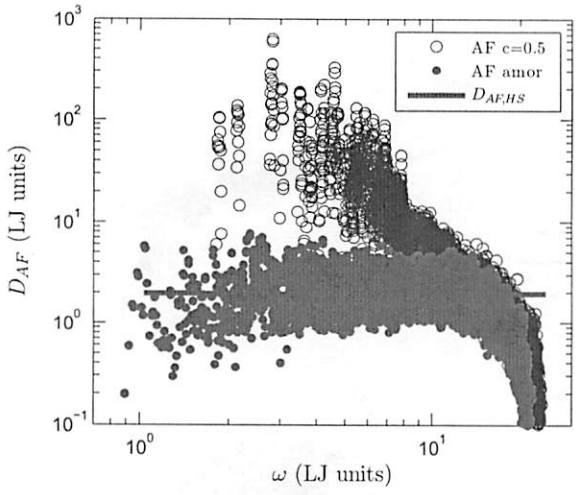


FIG. 7: AF theory predictions of disorded mode thermal diffusivities for LJ argon disorded lattice supercell and amorphous phase. The mode thermal diffusivities predicted for the disorded lattice supercell are all finite, except at the highest frequency where they tend to 0 as in the amorphous phase.

predicted by VC-NMD* is brought into agreement with GK by approximately 10% or less for all c. Combined with the high-scatter limit, the VC-NMD predicted thermal diffusivities are a fair representation of the explicitly disordered modes present in the MD simulation when

The VC-ALD method underpredicts the thermal conductivity of LJ argon alloys, where the underprediction is worst for $c = 0.05$, $k_{VC-ALD}/k_{GK} = 0.56$ (error bars are on the order of the large symbol sizes in Fig. 8). By applying the high-scatter limit adjustment VC-ALD*, the thermal conductivities are brought into marginally better agreement, worst for $c = 0.05$, $k_{VC-ALD^*}/k_{GK} = 0.65$.

The failure of the VC-ALD method can be demonstrated further by moving to higher temperature $T = 40$ K in Fig. 8 a. The beginning breakdown of the intrinsic phonon-phonon ($\tau_{p-p}(\kappa_\omega)$) scattering model can be observed for $c = 0.0$ at $T = 40$ K (Fig. 8 b), where ALD begins to overpredict compared to GK. This can be explained by the emerging importance of higher order ($n > 3$) n-phonon process at high temperatures.[34] While the VC-ALD method begins to overpredict for $c = 0.0$ at elevated temperature, it continues to underpredict for the alloys $c \geq 0.05$. In fact, the thermal conductivity predictions for VC-ALD are at or

- From the text, it is not totally clear why the correction works for VC-NMD but not for VC-ALD

- Can you do NMD at T=40K?

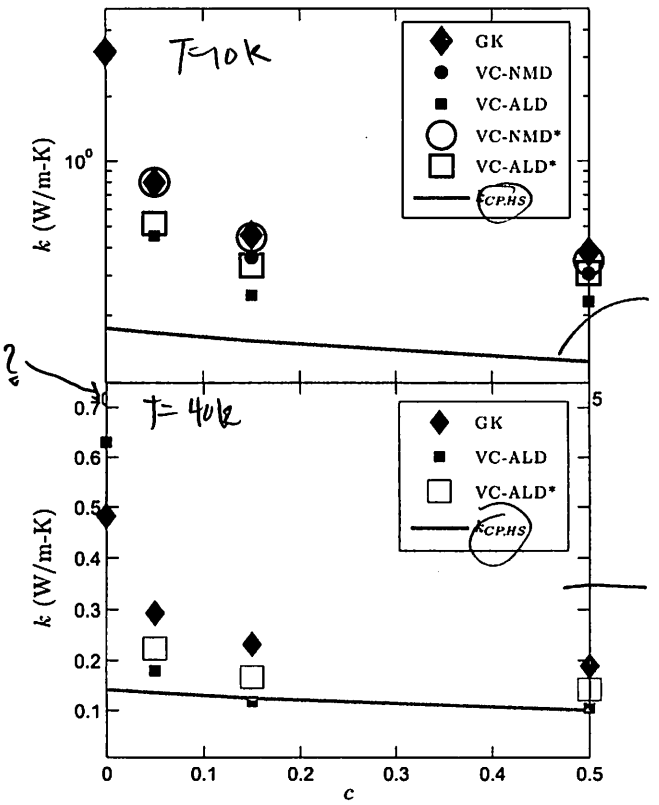


FIG. 8: (a) thermal conductivity predictions for LJ argon alloys at T=10K using the VC-NMD, VC-ALD, and GK methods. (b) thermal conductivity predictions at T=40K.

slightly below the high-scatter limit $k_{AF,HS}$.

explain what is plotted more carefully

The VC-ALD method fails to accurately predict the high-frequency mode thermal diffusivities for LJ argon alloys, which can be seen in the thermal conductivity spectrum Fig. 5 c.(cite) Since the group velocities are the same for VC-NMD and VC-ALD, this underprediction of the high-frequency thermal diffusivities is due to the underpediction of the high-frequency mode lifetimes (Fig. 5 a). At T=40 K, the thermal conductivity of the diffusivity adjusted VC-ALD* is only marginally improved compared to GK.

For SW silicon, the thermal conductivities predicted by VC-ALD and GK are in better agreement, even without the adjustment VC-ALD*. VC-ALD actually overpredicts by roughly 20% for $c \geq 0.05$ compared to GK. For SW silicon alloys, the VC-ALD* adjustment increases the result from VC-ALD by about 1% because the contribution from high-frequency modes is only a few percent (Fig. 6 c).

new section w/ small Si data

expand this discussion - maybe integrate with some of the text from section V?

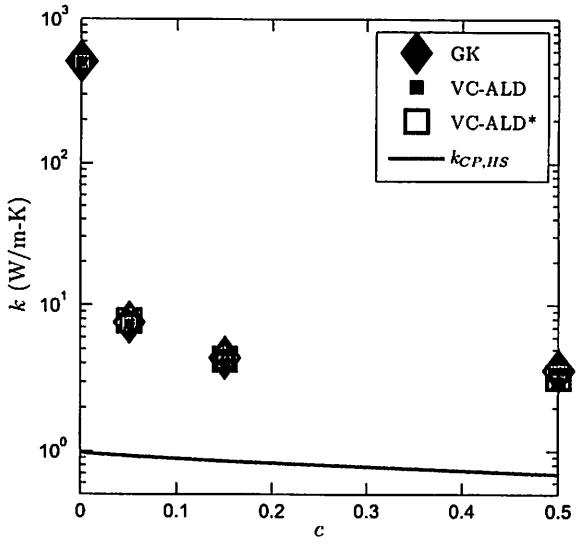


FIG. 9:

V. DISCUSSION

The LJ argon and SW silicon alloys studied in this work have different ranges of phonon frequencies, lifetimes, group velocities and total thermal conductivity. For bulk silicon(cite), the thermal conductivity is dominated by low-frequency modes(cite), which is also true for bulk and alloyed SW silicon (Fig. 6).(cite) For SW silicon, VC-ALD predicts thermal conductivities in reasonably good agreement with the explicitly disordered GK method (Fig. 9). For LJ argon, VC-ALD underpredicts the high-frequency phonon lifetimes and thermal diffusivities (Fig. 5), leading to an underprediction of thermal conductivity when compared to VC-NMD and GK (Section IV).

The results for SW silicon and LJ argon alloys suggest that the thermal modeling of ordered and disordered lattices can be separated into two broad groups: low- and high-frequency dominated materials.(cite) Materials dominated by low-frequency modes tend to have high thermal conductivities, which is due to their large group velocities and long lifetimes. (cite) These low-frequency modes follow closely the scalings predicted by the perturbative VC-ALD models, which are valid at low-frequencies.

LJ argon is a material dominated by high-frequency modes, even for the bulk (Fig. 5). This high-frequency range is where the perturbative VC-ALD models, and also where

the higher-order terms in the Tamura theory are predicted to be non-negligible (Section III C 2). While the high-order terms in the Tamura theory are also predicted to be non-negligible for SW silicon, this does not affect the thermal conductivty predictions signifcantly. Even if there was a discrepancy at high-frequency these modes are unimportant to thermal transport in SW silicon. This is also true for the thermal conductivity spectrum of SiGe alloys from first-princples predictions[6] and experimental measurements.[18? ? ,19] and first-principles predictions[5,6] this is also true for SiGe alloys and isotopic GaN. For example, the thermal conductivity of SiGe alloys exceeds the high-scatter limit by more than an order of magnitude at room temperature for all com- positions.

Resolution of the breakdown of the perturbative VC-ALD models for LJ argon is achieved by considering the thermal diffusivity of vibrational modes in the AF theory. A simple correction to the VC approximation can be made by considering a high-scatter limit for the mode specific thermal diffusivity (Section III D). This high-scatter limit is physically interpreted as vibrational modes propagating at the sound speed a distance of the lattice constant. However, the concept of a vibrational mean free path in a disordered system is only valid at low-frequencies.[43?] For disordered vibrations, the lifetime and thermal diffusivity are the fundamental quantities. The VC approximation underpredicts the mode thermal diffusivity due to disorder because the group velocities $v_g(\overset{\kappa}{\nu})$ can approach zero (Section), though this is a small effect compared to the underprediction of the lifetimes by VC-ALD.

For LJ argon, it is possible that the VC group velocities are an over-prediction for modes in a given interval of frequency, an effect which is compensated for by a small under-prediction of the lifetimes in the same interval of frequency when compared to Gamma-NMD (Fig. 4). The VC-NMD predicted mode lifetimes and thermal diffusivity adjusted VC-NMD*, predict thermal conducitivities in good agreement with the MD-based GK method. Based on the thermal conductivity predictions for VC-NMD* and the well-defined peaks in the structure factors (Fig. 3), the reduction of group velocities in disordered lattices due to zone folding seems to be an underprediction of the group velocity of moderate to high frequency modes.[26]

VI. SUMMARY

The concept of simple alloying is at the forefront of the effort to control or minimize the thermal conductivity of semiconducting and thermoeletric materials (cite SiGe nanoporous,

PbTe) Results in this work suggest that the lower limit for the vibrational mode thermal diffusivity in alloys with thermal conductivities near the high-scatter limit is $(1/3)v_s a$. Such materials include the thermoelectric PbTe, particularly at the high operating termperatures of thermoelectric energy generation.(cite) PbTe has a thermal conductivity in the mid-range of frequencies, placing it between SW silicon and LJ argon. The optical phonons in PbTe/PbSe alloys have been predicted to have group velocities as large as the acoustic branches, making PbTe/PbSe distinctly unique compared to SW silicon or LJ argon alloys.[7] It is not clear what role the high-scatter limit of the mode-specific thermal diffusivity has in predictions for PbTe/PbSe alloys.

The results in this work support the idea of a minimum thermal diffusivity for the vibrations in disordered lattices.(cite) Although this minimum thermal diffusivity is usually interpreted as a minimum mean free path, we find that concept is not necessary for interpreting the results. The VC approximation provides a computationally cheap framework, which is essential for expensive but experimentally accurate *ab initio* methods for predicting thermal conductivity.(cite) The high-scatter limit of thermal diffusivity is more useful for examing the thermal transport in alloys under the framework of the VC approximation. The fundamental quantity is the mode lifetimes and the group velocity is an approximation, and expressed together as thermal diffusivity they can be interpreted in the presence of disorder.

Acknowledgments

This work was supported in part by a grant of computer time from the DOD High Performance Computing Modernization Program at the US Army Engineer Research and Development Center. We thank Jivtesh Garg, Zhiting Tian, Davide Donadio, Asad Hasan and Craig Maloney for helpful discussions.

Appendix A: Computational Cost

The key to incorporating the effects of disorder explicitly are the use of a large disordered supercells (Section II C). However, the methods used in this work scale differently with the size of the supercell considered. The calculations in this work are trivially parallelizable except the MD simulations[7] and the eigenvalue solution of the Dynamical matrix.[25] Efficient

MD codes scale linearly with the number of atoms in the system N_a, making the GK method an efficient method for predicting thermal conductivity. However, the computational cost of using large supercells for MD simulation, particularly because of the large number of time steps required (on the order of $10^5 - 10^7$ depending on the system, time step used, etc (cite)), prohibit its use with typical *ab initio* methods such as plane-wave Density Functional Theory.(cite)

Both VC-NMD and VC-ALD require the eigenvalue solution of a Dynamical matrix of size $(3n, 3n)$ for each irreducible wavevector of the system size considered, which is negligible compared to the other caculations required for both of these methods.(cite) The Gamma-NMD (Section ??) and AF theory (Section III D) require the eigenvalue solution of a large Dynamical matrix $(3N_a, 3N_a)$, the solution of which scales as $(3N_a)^3$. The AF theory is limited to small supercells using ab initio calculations, making it difficult to asses finite-size effects.(cite)

Using the VC-ALD method, the symmetries of the system can be used to drastically reduce the required computations, permitting its use with ab initio methods.[9,34,44?] For VC-ALD, the calculation of the intrinsic phonon lifetimes $\tau_{p-p}(\substack{\kappa \\ \nu})$ scales as n^4,[34] making calcualtions for large unit cells challenging.(cite) Compared to the calculation of the intrinsic phonon lifetimes, calculation of the defect lifetimes $\tau_d(\substack{\kappa \\ \nu})$ (Eq.) is negligible.

Appendix B: NMD using Non-Exact Normal Modes

The NMD method requires the atomic positions and velocities from an MD simulation.(cite) The MD simulations are performed using the package LAMMPS.[?] The lengths of the MD simulations were longer than 10 times the longest phonon lifetime in the system. These can be estimated a priori from the VC-ALD predicted phonon lifetimes. For LJ argon and SW silicon, the simulations were run using time steps of $dt = 0.002$ LJ units and $dt = 0.0005fs$ for 2^20 and 2^22 time steps and the atomic trajectories were sampled every 2^8 and 2^4 time steps, respectively. Ensemble averaging was performed using 10 independent initial randomized velocity distributions.

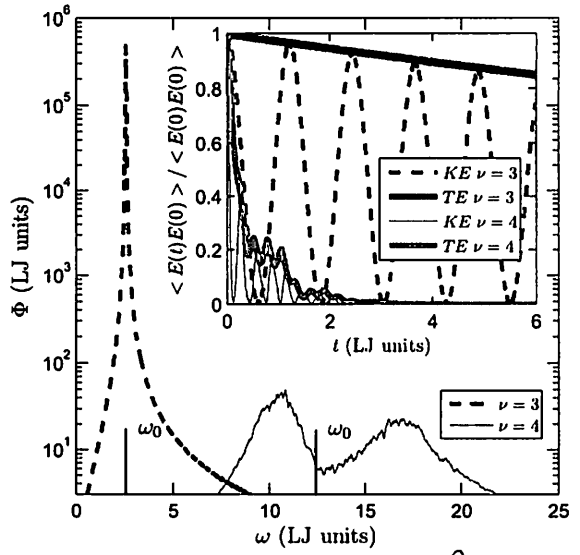
For a normal mode of a lattice supercell used for the MD simulations, the autocorrelation of the total and kinetic normal mode energy are damped exponentials with a decay time $\tau(\substack{\kappa \\ \nu})$, the kinetic energy autocorrelation with a cosinusodial oscillation frequency $2\omega(\substack{\kappa \\ \nu})$.(cite

joe) When using the VC normal modes to map the MD simulation trajectories for the explicitly disordered lattice supercells, the mode total and kinetic energy autocorrelation fucntions do not always follow ~~simple~~ functional forms. This can be illustrated by using spectral-NMD in the frequency domain, where artifacts such as multiple peaks in an isolated mode's energy spectrum (Φ) can be observed (see Fig).(cite) In the case of multiple peaks, the choice of which peak to fit to predict the phonon properties can be ambiguous. However, a lifetime can be predicted unambiguously using Eq. even with these multiple-peak artifacts, particularly because the autocorrelations are damped exponentially. This results is to be expected given that the atomic trajectories contain information about the lattice energy, which from general statistical physics principles will have exponential relaxation behavior in an equillibrium ensemble.[45? ,46]

These artifacts are not surprising given two considerations: 1) the MD simulations contain explicit disorder which incluences the atomic trajectories 2) the VC normal modes are not the exact normal modes and of the explicitly disordered system. Descrepencies have been observed previously when the exact normal modes of the system are not used.(cite SED) However, the lifetimes predicted using VC-NMD are in fairly good agreement with those calculated using Gamma-NMD (Fig. 4). Several studies have found good agreement for predictions of lifetimes and thermal conductivity using non-exact eigenvector mappings[10,47] in a wide-range of materials and phonon scattering conditions.[2,8,10,47,48] However, it is crucial that results using non-exact mappings are compared to as many alternative methods as possible. In this work, VC-NMD is compared to the other methods Gamma-NMD (Section ??), GK (Section ??), and VC-ALD (Section III D). It is important to remember that the VC normal modes are exact in the limit $c- > 0$. Use of the VC modes at large c pushes the limits of the approximation, but is useful for predicting an effective group velocity (Section III B).

Appendix C: Calculation of the Gamma Mode Structure Factors

To calculate $S^{L,T}(\frac{\kappa}{\omega})$ for a finite-size system, the delta function in Eq. (??) is broadened using a Lorentzian function with a full-width at half maximum $\Gamma_{FMHW} = \delta_{\omega,avg}$, where $\delta_{\omega,avg}$ is the average frequency spacing. Allen et al[31] demonstrated using a model of a-Si that the structure factor for large wavevector broadens so that the linewidth ~~Γ_{SF}~~ $> \omega$.? For



can we use a different term?

FIG. 10: The spectral energy density Φ of two modes (polarizations $\nu = 3, 4$ at wavector [0.2 0 0]) calculated using VC-NMD for a mass disordered LJ FCC supercell ($N_0 = 8$ and $c = 0.5$). The VC dispersion-predicted peaks are labeled by ω_0. Inset: the same mode's energy (kinetic (KE) and total (TE)) autocorrleation functions. Note the additional harmonic effects in the KE and TE autoccorelation functions for $\nu = 4$ which are due to the double peaks in Φ. A mode lifetime can be extracted unambiguously using the integral of the TE autocorrelation function (Section ??).

the systems sizes studied, Γ_{SF} scale with the brodening factor Γ_{FMHW} for all peaks except those at high frequencies.

For the range of broadening factors considered ($\Gamma_{FMHW} = \delta_{\omega,avg}$ to $50\delta_{\omega,avg}$) the linewidths extracted for all c generally satisfy $\Gamma_{SF} > \omega$. For all broadening factors, the linewidths (inverse lifetimes, $\tau_{SF} = 1/2\Gamma_{SF}$) at high frequency are in better agreement with the lifetimes predicted by VC-NMD rather than VC-ALD, where generally $\tau > 2\pi/\omega$ (IR limit, Fig. 4).[?] This gives more justification for the use of the VC predicted group velcoities for both VC-NMD and VC-ALD, even for large wavevector and c.

is it worth discussing this point? I feel that it is a digression from the rest of work

In general, the polarization of the eigenvectors $e(\substack{\kappa\ b \\ \nu\ \alpha})$ will not be purely transverse or logitudinal along the reciprocal directions. Even for the simple LJ argon system, this can make it difficult to uniquely identify then different polarizations with the various peaks in the structure factors. For SW silicon, similar good agreement can be seen along the high symmetry directions for the acoustic branches, while the optical modes and more complicated

random

polarizations are too difficult to identify in an automated way. In general, the acoustic branches can be identified, provided they are well separated in frequency from any optical branches.[47?]

Appendix D: Finite Simulation-Size Scaling for Thermal Conductivity

To predict a bulk thermal conductivity, extrapolation is used by the following finite size scaling $1/k \propto 1/N_0$. For VC-NMD and VC-ALD, the validity of the finite-size scaling requires the low frequency modes in the finite system to be dominated by intrinsic scattering ($\tau(\overset{\kappa}{\nu}) \propto \omega(\overset{\kappa}{\nu})^{-2}$) and follow the Debye approximation with respect to $v_{g,\mathrm{n}}$ and DOS $D(\omega(\overset{\kappa}{\nu}))$.[8,9] For LJ argon, this requirement is satisfied for modest system sizes (for $N_0 = 6$ to 12) so that both VC-NMD and VC-ALD predictions can be extrapolated to a bulk value. For SW silicon, the thermal conductivity is dominated by low-frequency modes (Fig. 6). Becasue of this, large system sizes (up to $N_0 = 42$) are needed to satsify the extrpaolation requirements and only VC-ALD can be used.(cite) This demonstrates the computational efficiency of the VC-ALD method which is necessary when computationally expensive ab initio methods are used (Section).[5,6,9?]

System sizes of up to $N_0 = 38$ are required to predict converged thermal conductivity of SW silicon alloys. For Si modeled using the Tersoff potential, system sizes of up to 64000 atoms are required to observe converged values of thermal conductivity using the GK method.[?] We find that similar system sizes are also required for

For the GK method, smaller system sizes $N_0 \leq 12$ are used for the finite size extrapolation for LJ argon and SW silicon . The validity of this result can be explained in terms of a combination of effects which are specific to the MD simulations.[9] In fact, for $c = 0$ the GK results are independent of system size for $N_0 = 4$ to $N_0 = 12$ for LJ argon.

No URL's in these entries

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[49] For the disordered lattices studied in this work for $c \leq 0.15$, the predicted k_{AF} is strongly system size dependent, indicating this diverging behavior. For LJ argon alloys at $c = 0.5$, the divergence with system size is small for the range of system size studied ($N_0 = 4$ to $N_0 = 12$), where $k_{AF}/k_{GK} = 0.93$ for $N_0 = 12$. ††

[50] For a finite system, the AF theory requires a broadening in frequency to predict the mode-specific thermal diffusivities. We use a Lorentzian broadening with a width of $\delta_{\omega,avg}$, see Section ??. ‡‡

[51] Analysis of these high-frequency modes shows that they are locons, modes which are spatially

needed?

localized in the Anderson sense. Feldman et al showed that the thermal diffusivities in a-Si show a sharp breakpoint at the on- set of localized states, where it tends to zero exponentially. The correct length scale associated with these modes is the mode correlation length(cite), whose uppper bound is the size of the simulation domain. §§

[52] The overprediction of thermal conductivity by VC-ALD may be related to the role of disorder in the ALD calculation.[6,34] While Garg et al found an overprediction of VC-ALD compared to experiment by a factor of two, the overprediction we observe in this work for SW silicon alloys is not as drastic.

This point should be in main text