

Evaluation of the Virtual Crystal Approximation for Predicting Alloy Phonon Properties and Thermal Conductivity

Jason M. Larkin¹ and A. J. H. McGaughey^{1,*}

¹*Department of Mechanical Engineering*

Carnegie Mellon University

Pittsburgh, PA 15213

(Dated: February 26, 2013)

Abstract

The virtual crystal approximation for mass disorder is evaluated by examining two model alloy systems: Lennard-Jones argon and Stillinger-Weber silicon. In both cases the perfect crystal is alloyed with a heavier mass species up to equal concentration and phonon frequencies, lifetimes and group velocities and thermal conductivity are predicted. These two alloy systems have different ranges of phonon frequencies, lifetimes. For Stillinger-Weber silicon, the virtual crystal approximation predicts phonon properties and thermal conductivity in reasonably good agreement with molecular dynamics-based methods. For Lennard-Jones argon, the virtual crystal approximation underpredicts the high-frequency phonon lifetimes, leading to an underpredicting of its thermal conductivity. Resolution of this underprediction is achieved by considering methods that treat the disorder explicitly.

Disordered lattices are a subgroup of disordered materials where the atomic positions follow a lattice structure but the constituent species are spatially random. Examples include isotopic solids, where the species have the same electronic structure but small mass variations, and alloys, where two distinct species are present.

I. INTRODUCTION

Disordered materials (i.e., alloys and amorphous solids) are used in applications ranging from semiconducting devices to thermally insulating barriers due to their low thermal conductivities. (cite) An alloy is a lattice composed of multiple species which are spatially random (e.g. isotopic germanium).¹ Alloying (creating a disordered lattice) remains an effective method to reduce the thermal conductivity while maintaining good electrical transport properties, (cite) For example, reducing the thermal conductivity of thermoelectric materials can improve the efficiency of thermoelectric devices.²⁻⁴

-alloys are also useful for their mechanical properties

Theoretical predictions for the thermal conductivity of disordered lattices dates back to the work of Abeles, who showed that mass and strain disorder dominate the thermal resistivity of Si_{0.5}Ge_{0.5} and (Ga,In)As alloys, respectively.⁵ The Abeles theory assumed that the vibrations in the alloys were phonons, delocalized propagating modes, whose properties were computed assuming the disorder was a perturbation. Except for low-frequency (long-wavelength) modes, it is not clear which vibrational modes are phonon-like in alloys with arbitrary disorder.

Side → mass (Ga, In) - As - Ge

(try not to use conductivity and resistivity in same sentence)

Does Abeles = the 2.

In the case of dielectrics, almost all of the heat is conducted by the vibrational modes of the system. Understanding how these vibrations contribute to thermal transport is crucial for predicting the thermal conductivity of ordered and disordered lattices.

Accurately predicting the thermal conductivity of a dielectric or semiconducting material requires the properties from the full spectrum of vibrational modes.⁶⁻⁸ Accurate predictions of these properties for bulk systems can be made with anharmonic lattice dynamics (ALD) theory using *ab initio* calculations.^{4,9-14} However, computational costs limit the size of computational cells in *ab initio* calculations to be less than 100 atoms, making it difficult to explicitly incorporate the effects of disorder.^{4,10,11,15,16}

Disorder is typically included using the virtual crystal (VC) approximation, where the anharmonic lattice dynamics (ALD), and the VC-ALD approach predicted phonon mode frequencies, lifetimes and group velocities of defected materials with relatively large (order 100 W/m-K^{10,11}) and small (order 1 W/m-K⁴) thermal conductivities that compare well with experimental measurements.

to p2 (*)

Under the VC approximation, the disordered solid is replaced with a perfect virtual crystal with properties equivalent to an averaging over the disorder (e.g., mass or bond strength).⁵

The use of ALD with the VC approximation (referred to herein as VC ALD) can base all

disordered lattice < Boron solid alloy

-include some mention of Tamura theory in introduction

We will refer to this approach as VC-ALD.

(i.e., all vibrational modes are phonons)

are performed the phonon-phonon and calculations on a small unit cell with averaged properties and treat the effects of intrinsic phonon- and disorder scattering as perturbations.^{1,4,5,18} No comprehensive study has been performed self-consistently to assess the applicability of this perturbative approach for a range of disorder (varying alloy concentrations, Section II B) using multiple predictive methods and test systems to test the self-consistency of the VC approximation.

The goal of this ^{objective} study is to investigate the use of the VC approximation for predicting vibrational mode properties and thermal conductivity of alloys by a detailed comparison of three predictive methods: (i) Molecular Dynamics (MD)-based normal mode decomposition (NMD) Section III C 1), (ii) MD-based Green-Kubo (GK) Section IV), and (iii) VC-ALD (Section III C 2). ^{Dis} Using computationally cheap empirical potentials for argon¹⁷ and silicon¹⁸ ^{(Stillm34-helm (sw))} we study the effects of disorder explicitly. ^{can} For both LJ argon and SW silicon, the perfect lattice is disordered with a heavier mass species up to equal concentration ($c = 0.5$), spanning a range of perturbative to large disorder. By spanning this range, the limits of the perturbative models are examined.

We predict the phonon mode properties of the VC: frequencies (Section III), group velocities (Section II B), and lifetimes (Section III C), and use them to predict thermal conductivity (Section IV). The breakdown of the perturbative VC-ALD method is examined in Section III C 2, and a simple guideline is suggested by the AF theory (Section III D), which predicts the thermal transport properties of the vibrations of the explicitly disordered lattices. ⁱⁿ ^{correction} ^{provided in}

The vibrational mode properties are then used to predict thermal conductivities in Section IV, allowing for a comparison to the predictions of the Green-Kubo method, where no assumptions about the nature of the thermal transport are required. etc... (Comment on I, II, VII)

II. THEORETICAL AND MODELING FORMULATION

A. Thermal Conductivity Prediction

To predict the thermal conductivity of disordered lattices, one begins with the theory for a perfect lattice. For a perfect lattice, all vibrational modes are phonon modes, which by definition are delocalized, propagating plane waves.⁶ Using the single-mode relaxation time approximation⁶ to solve the Boltzmann transport equation¹⁹ gives an expression for thermal conductivity in direction \hat{n} ,

$$\kappa_{ph,n} = \sum_{\kappa} \sum_{\nu} c_{ph}(\kappa) v_{g,n}^2(\nu) \tau(\kappa). \quad (1)$$

The remainder of the paper is organized as follows. In Section II, ... In Section III, the frequencies, group velocities, lifetimes, and diffusivities of the vibrational modes of the disordered lattices are predicted when the disorder is explicitly included and when it is treated as a perturbation under the VC approximation.

phonon molecule
the sum is over the 1st Brillouin zone,

Here, κ is the wavevector, and ν labels the phonon polarization. The phonon mode has frequency $\omega(\kappa)$, $c_{ph}(\kappa)$ is the phonon volumetric specific heat, $v_{g,n}(\kappa)$ is the component of the group velocity vector in direction \mathbf{n} , and $\tau(\kappa)$ is the phonon lifetime.

The SMRT approximation has been shown to be accurate for SiGe alloys and lower thermal conductivity materials, while larger conductivity materials such as GaN and Diamond require a full iterative solution to the BTE for more accurate predictions.

For the materials considered in this work, the lattices and the components of their thermal conductivity are cubically symmetric, so that we refer to k_{ph} as a scalar thermal conductivity. This is also true for the disordered lattices in the infinite-size limit. Since MD simulations are classical and obey Maxwell-Boltzmann statistics,

the volumetric specific heat is k_B/V per mode in the harmonic limit, where V is the system volume. This approximation has been shown to be valid for LJ argon and SW silicon and is used in this work so that direct comparisons can be made between methods.

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

diffusons $k_{AF} = \sum_{\text{modes}} \frac{k_B}{V} D_{AF}(\omega)$ only a function of ω^2 ? define ω (2)

where D_{AF} is the mode-specific thermal diffusivity of disordered vibrational modes defined at the wavevector [000].

In this high-scatter (HS) limit, the AF thermal conductivity prediction is

meaning what? $k_{AF,HS} = \frac{k_B}{V_b} b v_s a$ what is b ? (3)

where V_b is the volume of the unit cell, v_s is the branch-averaged sound speed, and a is the lattice constant.

The relative contribution of both phonons and diffusons to the total vibrational conductivity has been estimated to be approximately equal for a-Si, while earlier studies find that k_{ph} is substantially less. While studies have been performed on alloying the amorphous phase, the AF theory has not been applied to disordered lattices.

In the current study of disordered lattices, the AF theory predictions provide a lower limit for the contribution of a given vibrational mode within the computational framework of the VC approximation. This is essential given the computational cost of the AF theory (Section).

this comment is looking for far ahead - think about where else it will go.

low vs. high $k < 150 \text{ W/mK}$ can we provide a guideline?

put refs with materials, not at end

need some motivation here

relevant to this work? tied to your next paper

include later when you do the calculation?

Abeles theory = κ_c + Klemens-Callaway?

B. Virtual Crystal Approximation

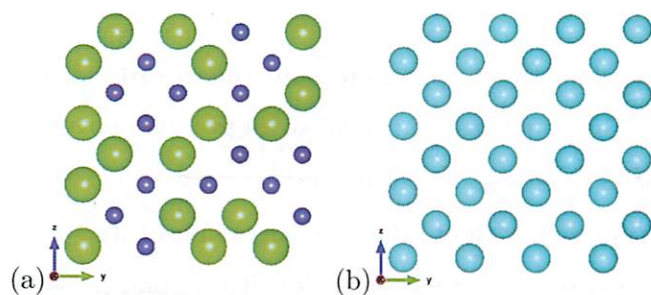
Under the ~~Virtual Crystal (VC)~~ approximation, the disordered solid is replaced with a perfect virtual crystal with properties equivalent to an averaging over the disorder (e.g., ^{atomic} mass and ^{spring} or bond strength).⁵ Abeles first introduced the idea of using a VC to replace a disordered one, ^{concept of} ~~predicting~~ ^{computing} the thermal conductivity of SiGe, (Ga,In)As and In(As,P) alloys using Klemens-Callaway theory.²⁶⁻²⁸ ~~and adjustable phenomenological fitting parameters to compare favorably with experimental results.~~ The Abeles theory is conceptually simple, treating both disorder and anharmonicity as perturbations and expressing the thermal conductivity as a closed-form analytical function of the bulk material properties.⁵ ~~While good agreement between the predictions and experimental measurements was found for SiGe and (Ga,In)As,~~ ^{with the use of phenomenological fitting parameters,} deviations from the Abeles theory were observed for In(As,P) alloys at large concentrations, which ^{were} attributed to the high mass ratio of 3.7 for InP.⁵ ^{which one?} ^{of which material}

While the Abeles theory ^{was developed} ~~is valid~~ for perturbative disorder, its ^{polarization} ~~use~~ leads to good agreement with ^{several} experimental and computational results for systems with a range of disorder. ^{It is important to note that} Cahill and co-workers found that ^{the thermal} conductivity reduction in dilute Ge-doped Si epitaxial layers can be explained by mass perturbative disorder.^{29,30} The effect of bond and mass disorder was investigated ^{sk} computationally by Skye and Schelling for SiGe³¹ ^{who also found} where it was shown that mass disorder is the dominant scattering mechanism. While the mass ratio is large (~~$m_{Ge}/m_{Si} = 2.6$~~), the overall disorder strength is determined by ^{sk} ~~both~~ ^{for example} the mass ratio, and the alloy concentration (Section III C 2). ^{not italic} As little as $6.2 \times 10^{19} \text{ cm}^{-3}$ Ge is enough to reduce ^{ratio (2.6)} the thermal conductivity of Si by ^{two} almost a factor of 2.²⁹ In the case of $(\text{Ni}_{0.55}\text{Pd}_{0.45})$, with ^{not italic} large mass disorder and concentration ^{ratio (2.6)} ~~($m_{Pd}/m_{Ni} \sim 2$)~~, good agreement is seen using the ^{the applicability of}

^{Abeles 2 theory} VC approach.³² Given these experimental results, it is unclear what limitations exist using the VC approach for arbitrary disorder ^{unclear}.

~~Computational results using the VC-ALD method for high thermal conductivity alloys show good to excellent agreement with experimental results for small and large concentrations.~~^{10,11} ^{OSIRIS} These VC-ALD computations use *ab initio* methods to predict the mode-specific phonon properties of the VC. Unlike the phenomenological Abeles theory, the VC-ALD ^{approach} predicts thermal conductivity by summing over the whole spectrum of mode-specific phonon ^{phonon} properties, where intrinsic and defect ^{between} scattering are treated as perturbations (Section). (cite) Lindsay and Broido found ^{quantity 5} good agreement ^{check spelling} with VC-ALD and experiment for

should we say ab initio or DFT or first principles?



you need to refer to this figure somewhere in the main text and discuss it

FIG. 1: (a) view of an explicitly disordered supercell of Si and "heavy" Si ([100] direction into the paper).³⁴ (b) view of the equivalent VC supercell with an average mass of the explicitly disordered Si and "heavy" Si supercell (b). Sphere size represents increasing mass only, no bond disorder is considered. In this work, calculations for LJ Ar and SW Si which use the VC approximation are based off of the conventional cubic unit cells (Section II C)

isotopically defected GaN with relatively small concentrations.¹⁰ Garg used (ab initio) calculations with VC-ALD to predict the thermal conductivity of SiGe alloys for all concentrations, obtaining excellent agreement with experiment.¹¹ Isotopically defected GaN and SiGe alloys have relatively large thermal conductivities. (cite) In particular, the thermal conductivity of SiGe alloys is significantly larger than ~~that of the~~ high scatter limit, which assumes that all vibrations scatter over a distance of the lattice spacing.²³ For both experiment and numerical modeling, VC predictions tend to be accurate when the thermal conductivity is significantly above the high-scatter limit.^{5,10,11,23,29,30,32}

A detailed study of low thermal conductivity materials PbTe¹² and PbTe/PbSe¹³ alloys made predictions for the perfect systems in fair agreement with experiment, but experimental results lack for the alloys.^{33?} Thus, there is a need to examine the perturbative approach of VC-ALD for large (SW silicon) and small (LJ argon) thermal conductivity alloys for perturbative and large disorder. While the computational studies discussed above were limited to VC-ALD because of ab initio calculation costs, we use computationally cheap empirical potentials to include disorder explicitly. It is a perturbation and to compare the predictions.

In this study, we will consider a low thermal conductivity alloy using the LJ potential and a high thermal conductivity alloy using the SW potential.

6 * This last paragraph is very important, but is similar to text in introduction. It may be worth the repetition. *

quantity
quantity

Eq. (3) or Cahill-Pohl?
Italic

Sound's figure

you should make this point around Eq. (3)
Is this your observation?
a bit ambiguous

Making it difficult to assess the validity of the ALD approach.
Comment on his scatter

there are some orphan sentences -
try to work on the flow

C. Calculation and Simulation Details

using the conventional unit cells

Perfect and ~~explicitly~~ disordered lattice supercells are generated ~~with atomic positions~~ ^{for} based on LJ argon's FCC ($n = 4$) and ^{SW} silicon's ~~diamond-FCC~~ ($n = 8$) ~~cubic conventional~~ unit cells, where n is the number of atoms in the unit cell. Supercells are built cubically with size N_0 , where N_0 refers to the number of ^{unit cell} repetitions of the unit cell in all ^{three} spatial directions. Supercells up to ~~size~~ $N_0 = 12$ (6096 atoms) are used for the LJ argon calculations. For SW silicon, $N_0 = 8$ (4096 atoms) is used for the MD-based NMD ^{calculations} methods, and $N_0 \leq 42$ (592704 atoms) ^{used for} MD-based GK and VC-ALD. The ALD calculations were performed ^{in the classical limit} using an in-house code ^{allow for comparison the} ~~with all classical expressions~~ to remain consistent with the classical MD-based methods ~~NMD and GK~~. The MD simulations were performed using LAMMPS.

comment on how even though you only introduce mass disorder, your systems are more like alloys than isotopic solids

Disorder is created by randomly specifying the masses of the atoms on the lattice. The composition of the lattices is labeled by $m_1^i \dots m_c^j$, where ⁽ⁱ⁾ $m^i = 1$ and $m^j = 3$ in LJ units for argon and ⁽ⁱⁱ⁾ $m^i = m_{Si}$ and $m^j = 2.6m_{Si}$ for SW silicon and "heavy silicon", ^{which has the} mass of germanium. Concentrations ^(c) of ~~var~~ 0.0, 0.05, 0.15 and 0.5 are used. For SW silicon, the lattice constant, ^{is} $a = 5.43 \text{ \AA}$ ^{a temperature of} is used for all calculations, which brings the GK thermal conductivity predictions at 300 K ^{36,37} into ^{compared to what?} better agreement with VC-ALD predictions for bulk SW silicon. ^{the} For LJ argon, supercells are built using the zero-pressure finite-temperature lattice constants, ^{at a temperature of 10 K is} which are $a = 1.556$ ($T = 10 \text{ K}$) and $a = 1.580$ ($T = 40 \text{ K}$) in LJ units. ^{give dimensional value} ^{needed?}

For LJ argon, the variation of lattice constant with composition is small and ignored. (cite)

Am The amorphous LJ phase, discussed in Section 0 was created by liquifying the crystal and instantly quenching by removing all kinetic energy. ^{energy of the} The resulting structure was then ^{energy} minimized and ^{from} annealed in an (NPT) ensemble at zero pressure and ^{a temperature of} ~~10 K~~ (cite lammps). The effective zero-pressure lattice constant of the amorphous phase at $T = 10 \text{ K}$, based on the atomic density, ^{xxx A} is slightly larger ($a = 1.585$). ^{define symbols}

At The MD simulations were performed using the package LAMMPS (cite lammps) by equilibrating in a NVT (constant number of atoms (N) volume (V) and temperature (T) ensemble. ^{are first} Data is then collected from simulations run in the NVE (constant number of atoms N , volume V and energy E) ensemble. (cite) For LJ argon, a cutoff of ~~2.50~~ ^{is used.} was used, where ~~3.4 \AA~~ ^{ten}. Statistical averaging is accomplished using 10 simulations with different initial velocities. MD simulation time steps of 4.285 and 0.5 fs were used for LJ argon and SW silicon. ^{how many dt?}

For the GK method, the heat current was computed every 10 time steps. It is important to

provide this detail later in section IV - too specific here

what is the cutoff scheme?

note that the same atomic trajectories are used for the NMD and GK methods.

The key to ^{explicitly} incorporating the effects of disorder ^{is to} ~~explicitly~~ are the use of large disordered supercells. ~~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 simulations⁴⁰ and the eigenvalue solution of the Dynamical matrix.⁴¹ Efficient MD codes scale linearly with the number of atoms in the system, N_a . However, The computational cost of using large supercells for MD simulation, particularly the large number of time steps required (on the order of $10^5 - 10^7$ depending on the system(cite)), prohibit its use with typical *ab initio* methods such as plane-wave Density Functional Theory.(cite) The Gamma-NMD and AF theory require the solution of a large Dynamical matrix of size $(3N_a)^2$, which limits the system sizes considered.

To predict a bulk thermal conductivity, extrapolation is used by the finite-size scaling $1/k(N_0) \propto 1/N_0$. For VC-NMD and VC-ALD, the validity of the finite-size scaling requires the low frequency modes in the system be dominated by intrinsic scattering $\left[\tau_{sc}^{-1} \propto \omega^{-2} \right]$ and follow the Debye approximation with respect to $v_{g,n}$ and $D(\omega)$.^{13,14} For LJ argon alloys, 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 alloys, the thermal conductivity is dominated by low-frequency modes, so that large system sizes are needed to satisfy the extrapolation requirements and only GK and VC-ALD can be used ($N_0 \leq 42$ in the present study, similar to the converged system sizes in³⁷). This highlights the efficiency of the VC-ALD method, which is necessary when computationally expensive *ab initio* methods are used (Section).^{4,10,11,14,42,43}

III. VIBRATIONAL MODE PROPERTIES IN ALLOY SYSTEMS

A. VC and Gamma DOS

In this section, we ^{begin to} examine the effect of explicit ^{by including} disorder by computing the ^{frequencies and} density of states $[DOS, D(\omega)]$ for vibrational modes of disordered lattice supercells and their equivalent VCs. The frequencies are computed using harmonic lattice dynamics calculations with the package GULP.⁴¹ For the VC, the allowed wavevectors are set by N_0 and each wavevector represents a finite volume of the crystal lattice's irreducible Brillouin zone (BZ).(cite mermin)

P = package?

and there are 12 polarization branches per wave vector

this comment seems more suited to lifetime section symmetry not important yet

where there are $12N_0^3$ polarization branches

The symmetry properties of the BZ define a set of irreducible wavevectors, which are used to average the mode properties of the reducible wavevectors. (cite)

For the disordered supercells, the only allowed wavevector is the gamma-point (i.e., $\kappa = 0$).

The DOS for the VC and the explicitly disordered supercells (referred to herein as Gamma) are shown in Fig. 2. The VC and Gamma DOS agree at low frequencies, where the Debye approximation predicts $DOS \propto \omega^2$. (cite)

The Debye approximation underpredicts the the DOS at moderate frequency, which is due to ~~the~~ non-linear dispersion. (cite Mermin)

The increasing lattice mass with increasing concentration for the VC reduces the frequencies. The increasing lattice mass for the Gamma modes also reduces the frequencies. The effect of explicit disorder is seen at high frequencies by a broadening and a shift of the DOS to higher frequencies because of the explicit use of light atoms in the supercell. Duda et al observed similar high-frequency broadening effects in model LJ alloys.⁴⁴

Similar agreement at low frequencies was found in *ab initio* predictions for $\text{Si}_c\text{Ge}_{1-c}$,¹¹ while Bouchard showed similar continuous behavior at low frequency for $\text{a-Si}_c\text{Ge}_{1-c}$.⁴⁵

Based on the DOS, the vibrational modes of the explicitly disordered supercells at low frequencies are phonon-like, while the broadening of the DOS at high-frequency indicate that the vibrational modes may differ significantly from the high-frequency VC phonon modes.

This behavior is further explained in the next three sections. What does the 'a' mean - amorphous?

write it out

you should be able to show a simple expression for the ω -shift

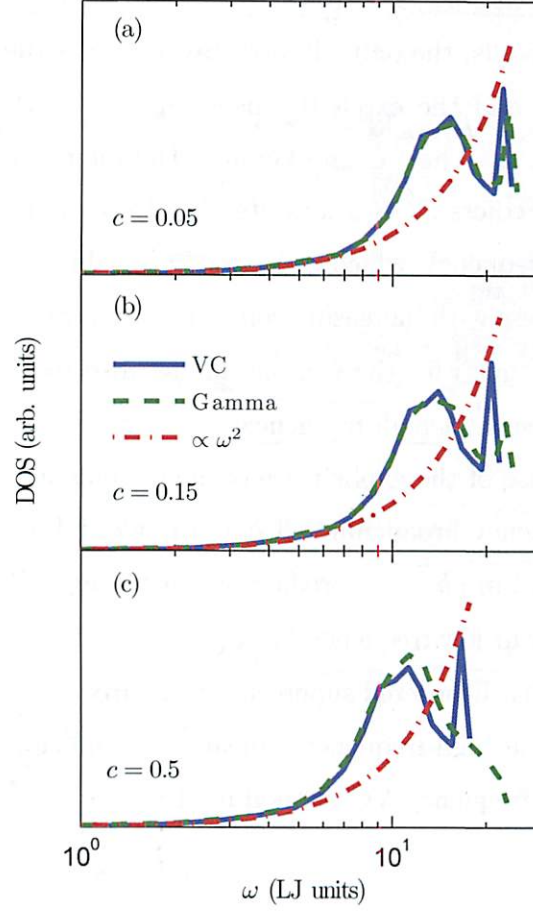


FIG. 2: Density of states (DOS) for modes calculated using the LJ FCC VC versus an explicitly mass disordered LJ FCC supercell (labeled Gamma) with varying mass concentration c . VC and Gamma show similar low frequency behavior for all c . For increasing c , the frequencies of both VC and Gamma decrease, while the high frequency DOS for Gamma spreads and reaches up to a higher maximum frequency because of the explicit disorder. The size of these supercells is $N_0 = 12$ (see Section II C).

B. Dispersion and Group Velocity

The group velocity vector in a VC is ^{returned} the gradient of the dispersion curves,

$$v_{g,n}(\kappa) = \frac{\partial \omega(\kappa)}{\partial \kappa}.$$

We calculate the group velocities for the VC using finite differences ^{on frequencies calculated from} and ~~quasi~~ harmonic lattice dynamics.⁴⁶

^{low frequency limit for the} Except for the three acoustic branches (2 transverse, 1 longitudinal sound speeds), ^{needed?} there is not an accepted method to predict the effective group velocity of a vibrational mode in a disordered system, although there have been attempts.^{23,24,44,47,48} In the Cahill-Pohl (CP) model, ^{for example,} the group velocity of all disordered modes is the sound speed, v_s .²³ Dispersion for a model disordered 1D system ^{however,} demonstrated the reduction of the frequency-dependent group velocities due to the zone-folding effect.⁴⁴

Calculating the structure factor of the supercell Gamma modes is a method to test for the plane-wave character of disordered modes at a particular wavevector and polarization.^{8,25} Feldman et al. ^{corresponding to the} used the structure factor to predict an effective dispersion for a model of a-Si, but did not predict group velocities.²⁵ Volz and Chen used the dynamic structure factor to predict the dispersion of crystalline SW Si using MD simulation.⁴⁹

The structure factor is defined as⁸

$$S^{L,T}(\omega) = \sum_{\nu} E^{L,T}(\kappa_{\nu}) \delta[\omega - \omega(\kappa_{\nu}^0)],$$

where E^T refers to ^{the} transverse polarization and is defined as

$$E^L(\kappa_{\nu}) = \left| \sum_{l,b} \hat{\kappa} \cdot e(\kappa_{\nu}^0 \frac{b}{a}) \exp[i\kappa \cdot r_0(l \frac{b}{a})] \right|^2$$

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

$$E^T(\kappa_{\nu}) = \left| \sum_{l,b} \hat{\kappa} \times e(\kappa_{\nu}^0 \frac{b}{a}) \exp[i\kappa \cdot r_0(l \frac{b}{a})] \right|^2. \quad (7)$$

Here, $r_0(l \frac{b}{a})$ refers to the atomic positions of the mass-disordered atoms in the supercell, which are still spatially ordered. Explicit disorder is accounted for in the mode frequencies $\omega(\kappa_{\nu}^0)$ and eigenvectors $e(\kappa_{\nu}^0 \frac{b}{a})$, which are calculated with $\kappa = 0$.

↑
you need to explain exactly what this term is

on frequencies calculated from

needed?

this comment needs more context

maybe use subscripts κ, Γ on κ 's?

what are b, a, l ?

Physically, $S^{L,T}(\omega)$ represents the frequency spectrum required to create a wavepacket with a well-defined wavevector and polarization.^{8,25} For a perfect lattice, the structure factor peaks are delta functions centered at the phonon mode frequencies, indicating they are pure plane-waves. ^{A sampling of the} The structure factors for LJ argon alloys are plotted in Fig. 3 for wavevectors along the [100] and [111] directions ^{in the No-10 systems}. With increasing disorder, the structure factor spreads in width, particularly at high frequencies ^{where} because the modes are no longer pure plane-waves.

From Fig. 3, an effective dispersion ^{curve} can be extracted by locating the peaks in the structure factors at neighboring VC wavevectors, ^{which are plotted in the middle panels} where the effects of polarization, virtual mass, and anisotropy ~~dispersion~~ can be observed. As the lattice VC mass becomes larger, the peaks in the structure factor shift to lower frequencies. The peaks in the structure factor are ^{at} slightly higher frequencies than the VC predicted frequencies by ^{at most} up to only ^{5%} ~~5%~~. Similarly, ^{2 new term} ~~good~~ agreement is found with the disordered SW silicon lattice supercells (not shown), ^{are} while the structure factors are more complicated because of the optical modes. Well-defined peaks at all wavevectors are ~~most likely~~ due to the lattice structure of the disordered systems ~~studied in this work~~. Typically, the structure factor for amorphous materials has well-defined peaks only for small wavevector.^{8,25} ^{larger}

Because of the good agreement between the VC ^{curves} predicted dispersion and the peaks in the structure factors from Fig. 3, we ^{will} use the group velocities predicted by the VC dispersion for both LJ argon and SW silicon ⁱⁿ with the VC-NMD and VC-ALD calculations for consistency and simplicity. We will examine the validity of this ~~choice of group velocity~~ ^{choice} in Section III D.