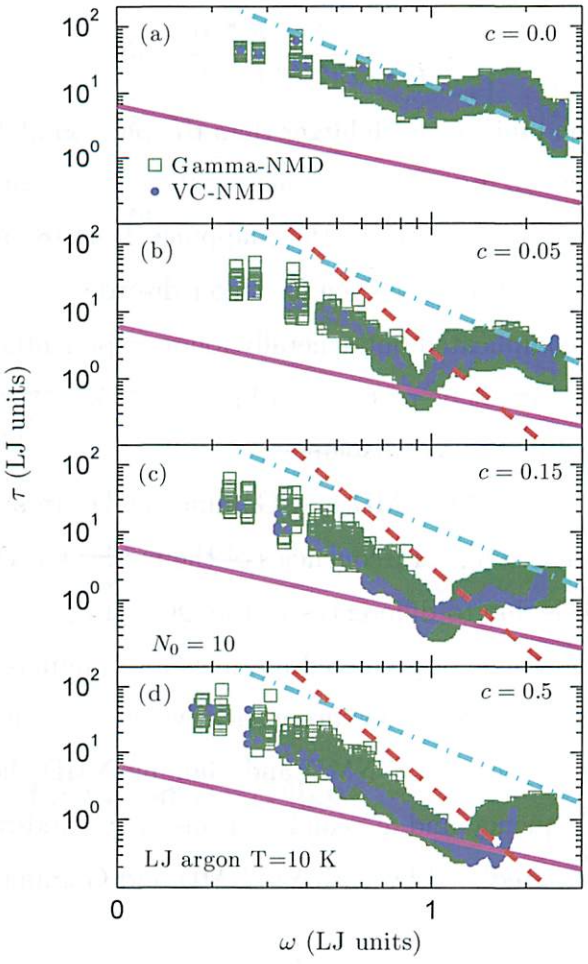

FIG. 4: Lifetimes predicted using VC-NMD and Gamma NMD from MD simulations of mass disordered lattice supercells (Section III C 1). Both ω^{-2} and ω^{-4} scalings can be observed at low frequencies, which are predicted by the peturbative models used for VC-ALD (Section III C 2). For both VC-NMD and Gamma NMD, most mode lifetimes are greater than the Ioffe-Regel limit $\tau = 2\pi/\omega$.[51] While there is more "noise" in the Gamma-NMD data (Section III C 1), the lifetime magnitudes and trends agree well, an important consideration when comparing VC-NMD and VC-ALD in Fig. 5 .

frequencies and eigenvectors to map the atomic trajectories from the fully anharmonic MD simulations, which has been shown to be valid ~~at the low~~ temperatures.[35] Based on the good agreement with Gamma-NMD, the lifetimes predicted by VC-NMD are used along with the VC predicted group velocities to predict thermal conductivity in Section IV.

The normal ~~vibrational~~ mode lifetime is predicted using

$$\tau(\kappa_\nu) = \int_0^{t^*} \frac{< E(\kappa_\nu;t)\, E(\kappa_\nu;0) >}{< E(\kappa_\nu;0)\, E(\kappa_\nu;0) >} dt, \tag{11}$$

where the upper integration limit is much larger than the phonon lifetime.(cite) For normal modes of the lattice supercell, Eq. (11) is exact, but becomes an approximation when using non-exact normal modes to perform the mappings, Eqs (8) and (8). An effective lifetime can be predicted using Eq. (11), even for larger disorder ($c = 0.5$), where the energy autocorrelations are more complicated but generally follow exponential decay (see Appendix A). ~~The phonon frequency cannot be predicted using Eq. (11), so the mode frequencies are taken to be those predicted by the VC dispersion.~~

The lifetimes predicted using VC-NMD and Gamma-NMD are shown in Fig. 4 for LJ argon alloys at 10 K. The range of frequencies ~~of the modes~~ for VC-NMD and Gamma-NMD differ slightly, ~~which is~~ due to differences in the DOS (Fig. 2). For small intervals of frequency, there ~~are~~ a wider range of predicted lifetimes for Gamma-NMD. This is because there is no symmetry averaging of the mode properties, which is possible for the VC ~~(Section III A)~~. Lifetimes predicted by both VC-NMD and Gamma-NMD show scaling ~~of τ with frequency of~~ ω^{-2} at low frequency and ω^{-4} and even faster for mid-range frequencies ~~(Fig. 4). In general,~~ the lifetimes predicted by both VC-NMD and Gamma-NMD are larger than the Ioffe-Regel (IR) limit,[51]

$$\tau = \frac{2\pi}{\omega}. \tag{12}$$

The physical interpretation of the IR limit is that of a mode which scatters in a time equal to its oscillation period, ~~which seems to be~~ a good lower-limit for the lifetimes predicted by VC-NMD and Gamma-NMD for LJ argon (Fig. 4) and VC-NMD for SW silicon [Fig. 8(a)]. ~~The behavior~~ at the highest frequencies, ~~where τ constant, is seen~~ for both VC-NMD and Gamma-NMD, except at $c = 0.5$ for VC-NMD. ~~Since the existence of this characteristic (thought not exactly minimum) lifetime for LJ argon is demonstrated by both VC-NMD and Gamma-NMD, it is physically meaningful.~~ There is, however, no theoretical prediction of this high-frequency behavior ~~of the mode lifetime~~.[23,52,53]

Overall, good agreement is seen in the predicted lifetimes from VC-NMD and Gamma-NMD both in magnitude and trends. The use of the VC normal modes is an approximation ~~which~~ becomes worse as the concentration is increased (Appendix A). The only approximation associated with Gamma-NMD is the use of the harmonic lattice dynamics predicted

C. Lifetimes

1. *From VC-NMD and Gamma-NMD*

Once the group velocities are predicted using the VC dispersion, the phonon mode lifetimes are required to predict the thermal conductivity using Eq. (1). As an alternative to the VC-ALD models for predicting phonon lifetimes, which are discussed in the next section, we first use the normal mode decomposition (NMD) method.[35,50] In NMD, the atomic trajectories (positions and velocities) of atoms in an MD simulation are first mapped onto the vibrational normal mode coordinates, (cite)

$$q(^{\kappa}_{\nu};t) = \sum_{\alpha,b,l}^{3,n,N} \sqrt{\frac{m_b}{N}} u_\alpha(^{l}_{b};t)\, e^*(^{\kappa\ b}_{\nu\ \alpha}) \exp[i\boldsymbol{\kappa}\cdot \mathbf{r}_0(^{l}_{0})] \tag{8}$$

and

$$\dot{q}(^{\kappa}_{\nu};t) = \sum_{\alpha,b,l}^{3,n,N} \sqrt{\frac{m_b}{N}} \dot{u}_\alpha(^{l}_{b};t)\, e^*(^{\kappa\ b}_{\nu\ \alpha}) \exp[i\boldsymbol{\kappa}\cdot \mathbf{r}_0(^{l}_{0})], \tag{9}$$

where $\mathbf{r}_0(^{l}_{0})$ are the equilibrium positions of the atoms in the lth unit cell of the lattice supercell under the VC approximation. The total energy of a given vibrational mode is

$$E(^{\kappa}_{\nu};t) = \frac{\omega(^{\kappa}_{\nu})^2}{2} q(^{\kappa}_{\nu};t)^*\, q(^{\kappa}_{\nu};t) + \frac{1}{2}\dot{q}(^{\kappa}_{\nu};t)^*\, \dot{q}(^{\kappa}_{\nu};t) \tag{10}$$

We perform NMD using the frequencies and eigenvectors from both the VC $\left[\omega(^{\kappa}_{\nu}),\ e(^{\kappa\ b}_{\nu\ \alpha})\right]$ and the Gamma supercell $\left[\omega(^{\kappa=0}_{\nu}),\ e(^{\kappa=0\ b}_{\nu\ \alpha})\right]$. The trajectories from these MD simulations are also used in the GK method (Section IV). ~~For the NMD method (Section III C 1), the atomic positions and velocities were sampled at a rate dictated by the highest vibrational frequencies in the system, which can be estimated from harmonic lattice dynamics calculations (Section III A).~~

~~The MD~~ simulations are performed ~~using the package LAMMPS.~~[40] ~~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 $dt = 0.0005$ fs~~ 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.

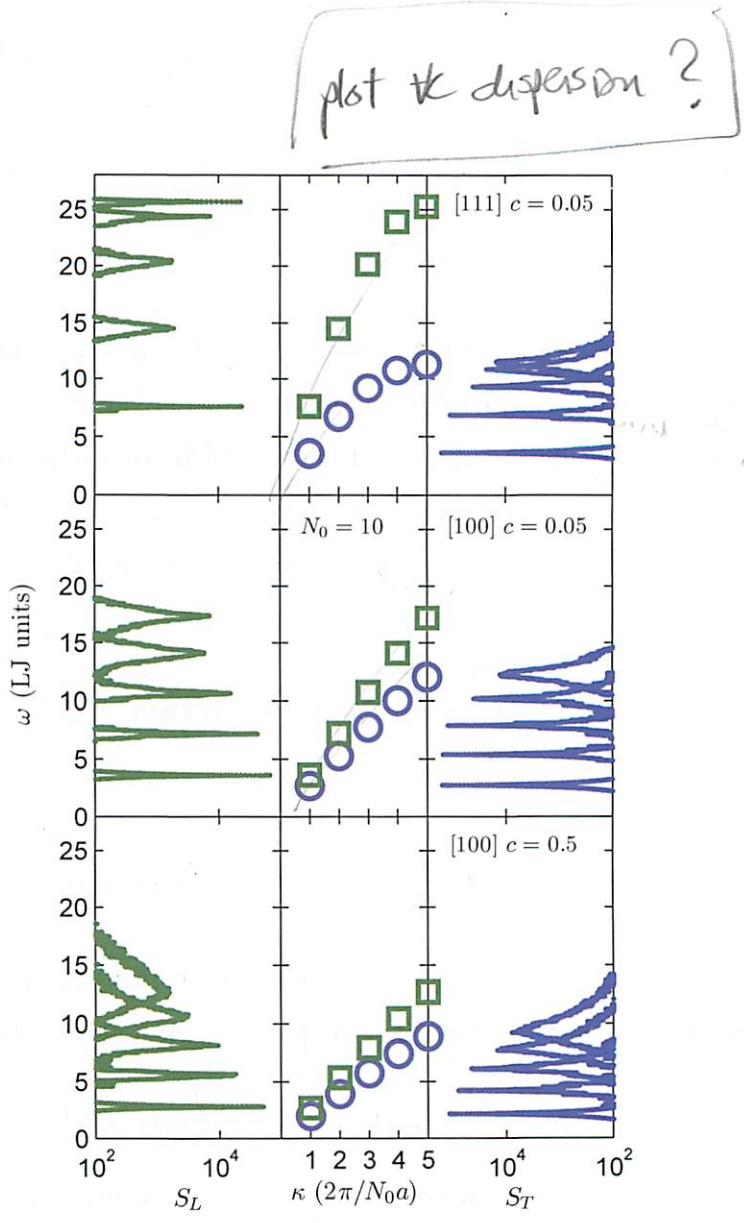

FIG. 3: Left and Right Panels: The structure factor for logitudinal (S_L) and transverse (S_T) polarizations along high symmetry directions ([100], [110] where $\kappa = \pi/a$[100] and a is the lattice constant) of the mass disordered LJ argon supercells ($N_0 = 10$,$c = 0.05, 0.5$). For increasing mass disorder c, there is a decrease in the center of the peaks and an increase in the peak linewdiths. Center Panel: The VC predicted dispersion at the same wavectors used to calculate $S_{L,T}$.