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Phonon Lifetimes: A Dependency on Size for Nano Silicon and Nano Germanium

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Abstract

Molecular dynamics (MD) was used to calculate the phonon dispersion relations of silicon (Si) and germanium (Ge) nanowires, from which the phonon lifetimes were deduced, and investigate the diameter dependency. Structures ranging ~ 2 – 4 nm thick were investigated, as well as a bulk counterpart for both elements. This study presents the results of using a simplified method to determine the phonon lifetimes of nanoscale systems.

1. Introduction

Predicting phonon behavior is key to understanding the reduction of thermal conductivity in nanoscale sized semiconductors. Phonon lifetimes characterize the intervals between scattering events, making this property a heavily researched field. However, the difficulty lies with accurately determining the phonon lifetimes. Pump-probe spectroscopy [1] and equilibrium MD [2] are examples of widely used techniques for the experimental and computational methods respectively. Thomas et al. [3] reported a simplified method for determining the phonon properties of carbon nanotubes using the spectral energy density (SED), finding good agreement with lattice dynamics data. Larkin et al. [4] claimed this method to be invalid as the total SED cannot be produced but having only tested bulk and large systems. A more recent study [5] applied this method to 1D atomic chains and found the SED resolution to increase with simulation domain but did not claim the results to be invalid. Nevertheless, less focus has been placed on nanoscale Si and Ge for this fitting method and as such, forms the theme of this study.

2. Calculation Methods

Phonon Dispersion Relation

Using MD, the phonon dispersion relations were obtained from the Fourier transform via SED. SED is the energy density per frequency space and is calculated by applying the atomic velocities to obtain the normal modes of vibration. The average kinetic energies of the normal modes are deduced and converted from the time domain to the frequency domain, the sum of which, is then averaged over the total simulation size to give the SED. For a phonon mode ($\boldsymbol{\kappa}$, ω), where $\boldsymbol{\kappa}$ is the wavevector and ω is the frequency, SED, Φ, is given by [3],

$$\Phi(\boldsymbol{\kappa},\omega)=\frac{1}{4\pi\tau_0 N_T}\sum_a\sum_b^B m_b\left|\int_0^{\tau_0}\sum_{n_{x,y,z}}^{N_T}\dot{u}_a\binom{n_{x,y,z}}{b};t\right)\times exp\left[i\boldsymbol{\kappa}\cdot\boldsymbol{r}\binom{n_{x,y,z}}{0}-i\omega t\right]dt\right|^2. \quad (1)$$

where N_T is the total number of unit cells in the lattice, $\dot{u}_a$ is the displacement in direction a of atom b, with atomic mass m_b, τ_0 the phonon lifetime, the unit cell $n_{x,y,z}$, t for time, r for the equilibrium positions of unit cell $n_{x,y,z}$.

Square cross-sectional nanowire (NW) structures of diameters 2.17, 3.26 and 4.34 nm with a length of 16.30 nm were tested for Si. Figs. 1(a) and 1(b) show the phonon dispersion relation of Si NW with a diameter of 3.26 nm, and the atomistic model respectively. Ge with a length of 16.97 nm was also tested, diameter sizes consisting of 2.26, 3.39 and 4.53 nm, as well as a bulk version for both elements. For the nanowires, a periodic boundary condition was applied in the longitudinal direction and for the bulk, the boundaries were applied in the x,y,z direction. The interatomic potential parameters used for the Si-Si and Ge-Ge pairs were developed by Tomita et al. [6]

Phonon Lifetime

For a given wavevector, the SED can be plotted as a function of frequency (termed in this extended abstract as SED-frequency plots). The lifetimes can then be deduced from the linewidths via the application of the Lorentzian function given by [3],

$$\Phi(\boldsymbol{\kappa},\omega)=\frac{I}{1+[(\omega-\omega_c/\gamma]^2} \quad (2)$$

where I is the peak magnitude, ω_c is the frequency at the peak center and γ is the half-width half-maximum. The phonon lifetime, τ, can then be obtained via τ=1/2γ [3]. Median and mean filtering was also applied to these plots in order to assist with deducing which peaks were of significance, and which peak ranges were to be classed as background noise or residual vibration. Corresponding to Figs. 1(a) and 1(b), Fig 1(c) shows the SED-frequency plot of the 3.26 nm diameter Si NW, with the raw data depicted in blue, the filtered data as the dashed black plot and the Lorentizan fitting as the orange plot.

3. Results and Discussion

Phonon Lifetimes

Fig. 2(a) shows the dependency of size on phonon lifetimes for Si NW along wavevectors k_x=0.3 and k_x=0.5. There is reasonable agreement with the NW lifetimes simulated by Donadio et al. [2] The lifetimes in Fig. 2(a) are similar across the diameters, exhibiting no definitive trend. Ge NW (Fig. 2(b)) exhibits similar behavior to the Si NW. There is no clear size dependency but viewing the results on a larger scale suggests the lifetimes will increase as the system reaches the bulk scale.

In addition, Fig. 3 in Ref. [1] further supports the observation that the size dependency is clearer when comparing

systems on a larger scale, but at a scale where the difference in diameters is within the 1 nm range, a trend cannot be deduced.

Larkin et al. [4] concluded that the method reported by Thomas et al. [3] is invalid, but only tested bulk and large systems. The results of this current study suggest that this method is suitable towards smaller nanoscale structures.

4. Conclusions

Si and Ge NW structures, as well as the bulk, were simulated to extract the phonon dispersion relations and phonon lifetimes using SED, a method reported by Thomas et al.[3] The bulk results indicated a higher resolution was needed, but the NWs produced results that are in reasonable agreement with literature. This method is suggested to be more suited towards nanostructures with 1 or 2 dimensions, rather than the bulk. The phonon lifetimes are largely influenced by the linewidths, consequently, the fitting procedures also. It cannot be concluded that there is a size dependency on the phonon lifetimes of these NWs as the difference between the diameters are too small. Testing diameters that differ greatly would assist with this conclusion.

Acknowledgements

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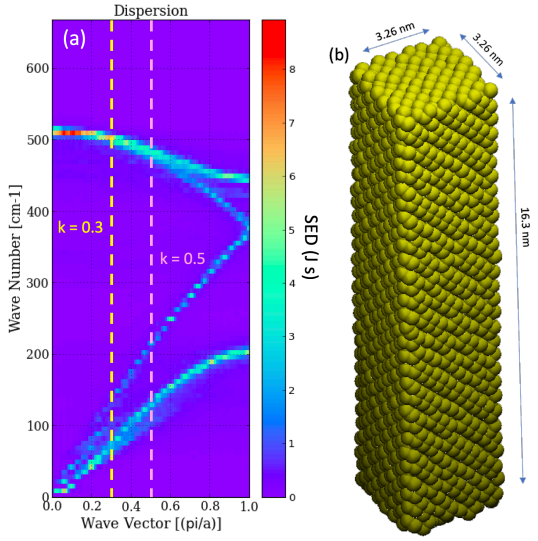

Fig. 1(a) Phonon dispersion relation for the Si NW with a 3.26 nm diameter. The wavevectors analyzed, k_x=0.3 and k_x=0.5, are indicated as dashed lines. The corresponding atomistic model of the 3.26 nm Si NW is shown in 1(b).

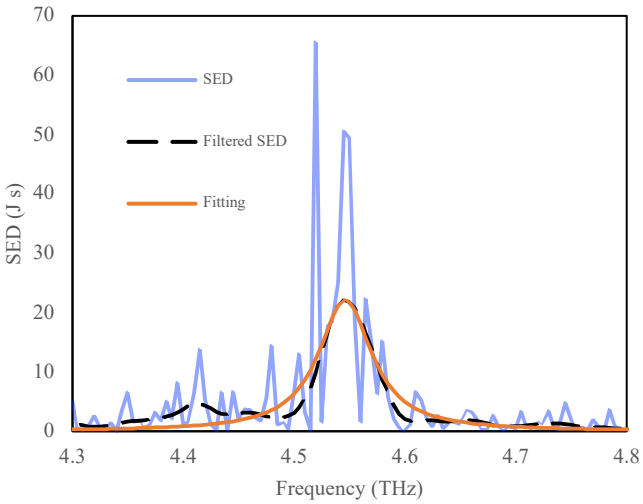

Fig. 1(c) SED-frequency plot for 3.26 nm Si NW, showing the SED unfiltered data as the blue plot, the filtered SED as the black dashed line and the Lorentzian fitting as the orange plot.

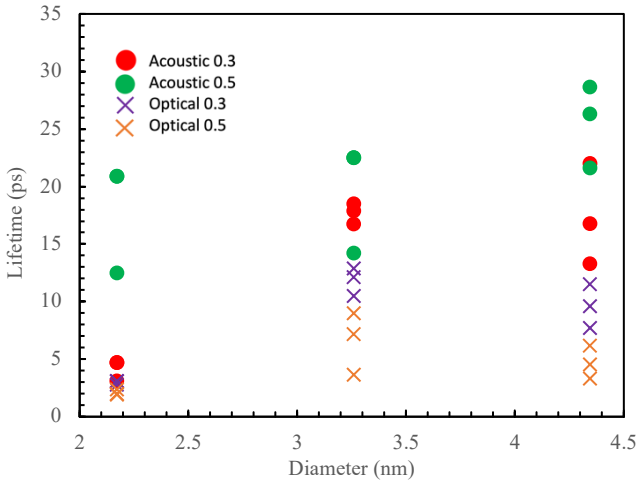

Fig. 2(a) Si NW phonon lifetimes as a function of diameter.

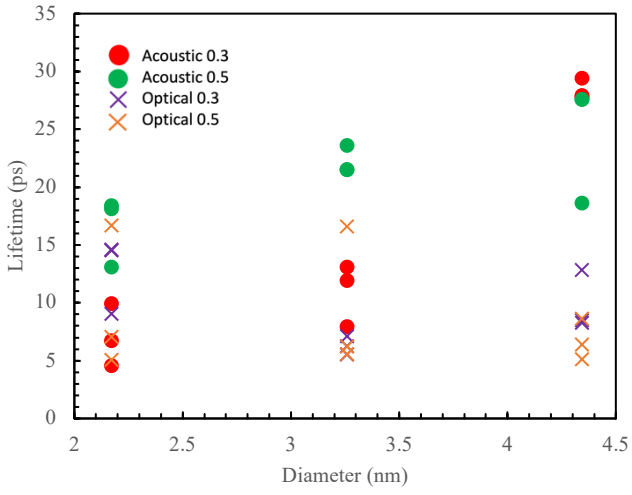

Fig. 2(b) Ge NW phonon lifetimes as a function of diameter.