First Principle Interatomic Force Constants for Thermal Conductivity Applications

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Abstract:

An accurate knowledge of heat transfer across semiconducting interfaces is critical to the performance of electronic devices. A phonon transport model capable of determining the thermal resistance at interfaces would provide better understanding of heat transfer issues in microelectronics materials, and also serve as a framework for designing new material interfaces. In this work, we examine phonon transport across a silicon-germanium interface using force constants based on first principles calculations. A density functional approach with a localized orbital basis set is used to determine the self-consistent charge and potential for bulk silicon, germanium, and a silicon-germanium interface. Once the equilibrium structure is determined, the interatomic force constants between atoms are extracted from a series of supercell calculations. With these interatomic force constants, the phonon dispersions for silicon (Si) and germanium (Ge) can be calculated and compared to experiment. The strength of the force constant as a function of distance between interacting atoms is also considered for the three systems. For bulk Si and Ge, we find that a large supercell (250 atoms) is required to generate phonon dispersions in good agreement with experiment. When plotted as a function of distance, the force constants generated between the displaced atom and its nearest neighbors (NN's) along the interface were relatively higher than the force constants between the systems containing only silicon or germanium. We will also discuss how these first principle interatomic force constants can be incorporated into a non-equilibrium Green's function approach to determine the interface thermal resistance.

Introduction:

Electronic devices that are made for consumer needs are expeditiously being created on the nanoscale [1]. As devices such as computer processors continue to be made smaller, the power density at semiconducting interfaces has increased dramatically as well. Interfaces are critical for removing heat from layers with a high power densities and thermal hot spots. Accurate modeling of the phonon movement across these semiconducting interfaces provides insight into the thermal resistance at these barriers. Building a phonon transport model capable of accurately calculating the thermal resistance at interfaces would provide greater understanding of heat transfer issues for electronic materials and help to design new materials that will optimize heat transfer. For this study we used the density functional theory program *Spanish Initiative for Electronic Simulations for Thousands of Atoms* (SIESTA) to determine the self-consistent charge and potential for bulk silicon and germanium, and a silicon-germanium interface [2]. The interatomic force constants calculated between atoms within the each lattice system were extracted and used to generate a phonon spectrum and incorporated in a Green's function to calculate the thermal conductance [1].

Procedure:

In order to accurately model the phonon transport across the silicon-germanium interface, the model needed to be tested for bulk silicon and bulk germanium lattices. First, the lattice constant for the bulk silicon system were determined by relaxing the atomic spacing and plotting the energy of the system as a function of volume. The volume versus total energy curve was used to determine the equilibrium lattice constant and associated bulk modulus. This was done for bulk germanium as well. The obtained lattice constants and bulk modulus compared well with experimental data. Once accurate lattice constants for bulk silicon and bulk germanium were found, the unit cell for the each system was created. The unit cell for silicon contained parameters such as the lattice constant, the type of atoms, and the number of atoms within the unit cell. Once the unit cell was created a program called fcbuild was used to create the super cell lattice. The program duplicated an individual unit cell in the x,y,z directions to create a larger lattice composed of many identical cells [3]. The density functional theory program SIESTA was then run on the supercells for bulk silicon and bulk germanium. SIESTA used a localized orbital set to describe wave function of the crystal structure being tested. The program calculated the self-consistent potential and self-consistent charge of the lattice being tested, made an initial guess on the wave function that describes the atomic movement within the lattice, and calculated the total energy of the system.

Next, SIESTA moved the central atoms within the lattice in six directions (positive and negative x,y,z) and calculated the band structures, total energy, and force constants between atoms with each atomic movement. Next, a program called *vibra* was used to determine the phonon spectrum based on the force constants generated from the SIESTA calculations [3]. This process was done for bulk silicon and bulk germanium lattices containing 54 atoms and 250 atoms. The phonon spectrum generated for these calculations were compared to experimental data. Once the model produced accurate phonon spectrums for the bulk silicon and bulk germanium systems, the silicon-germanium interface was then created [4]. The unit cell for the interface contained twelve layers; the first five were germanium, the middle two were silicon, and the next five were germanium.

Using SIESTA, the atomic positions near the interface were relaxed to their minimum force configuration. The program fcbuild was then run to create the large super cell containing 108 atoms. The same process as the bulk systems was done for the interface to calculate force constants and generate the phonon spectrum [5].

Results:

Shown in Figures 1 and 2 are the phonon spectrum for the bulk silicon and bulk germanium lattices. Figure 1 shows the theoretical phonon spectrum and experimental data for a 54 atom and 250 atom silicon lattices. Figure 2 shows the theoretical phonon spectrum and experimental data for a 54 atom and 250 atom germanium lattices. Figure 3 shows the calculated force constant plotted as a function of distance for bulk silicon and bulk germanium, while Figure 4 shows the force constant versus distance graph for the silicon-germanium interface.

Discussion/Conclusions:

It is shown from Figure 1 and Figure 2 that in order to provide accurate modeling of phonon movement for bulk silicon and bulk germanium using SIESTA, a supercell containing at least 250 atoms is necessary. This increase in accuracy is seen when the 54 bulk cases and 250 bulk cases are compared along the (-K symmetry line in the Brillouin zone. Along this line, the 250 atom silicon and germanium curves are in better agreement with the experimental data than the 54 atom curves [1,6]. The increase in accuracy makes sense considering when the density functional approach was implemented there was a larger sample size to approximate the phonon frequencies with their associative vectors for the 250 atomic supercell. Figure 3 shows force constants that were calculated form the density functional theory for a central atomic shift. The accuracy of the model is reaffirmed when looking at the force constants for the nearest neighbors (NN's) of the shifted atom for the bulk silicon and germanium systems. The force constant values for the nearest neighbors for silicon are higher than germanium which is in agreement with their bulk modulus values. Figure 4 shows the force constant versus distance graphs for an in-plane atomic shift and out-of-plane atomic shift for a silicon atom in the middle layer of the silicon-germanium interface. It is seen that for the NN's from the bottom graph of Figure 4 that the interface force constant values for the in-plane shift are very close to the force constant values for silicon. This makes sense considering that if an atom was shifted within the plane interactions with other silicon atoms would dominate. Consequently, there would be more variance in the force constants along the interface for an out of plane shift because there are more atomic interactions incorporated [6,7].

In conclusion, the model is now accurate enough to be tested on a larger silicon-germanium. In the near future the phonon dispersions and force constants for the current interface will be used in a Green's Function approach to calculate the thermal conductance. This will lead the model becoming more accurate in testing the thermal conductance across more complex interfaces [6,7].

References:

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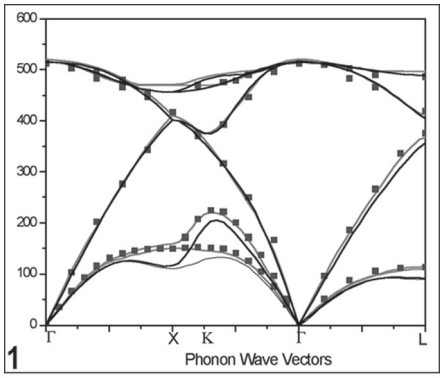

Figure 1: The phonon spectrum for bulk silicon. The black line represents the 54 atom system, while the red line represents the 250 atom system. The blue squares are the experimental data.

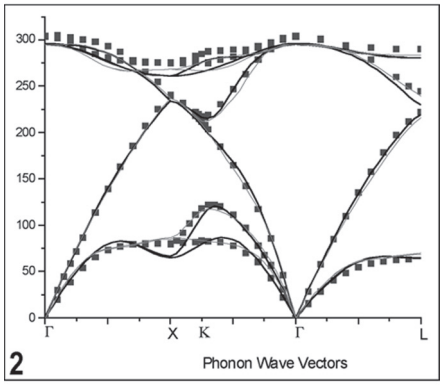

Figure 2: The phonon spectrum for bulk germanium. The black line represents the 54 atom system, while the red line represents the 250 atom system. The blue squares are the experimental data.

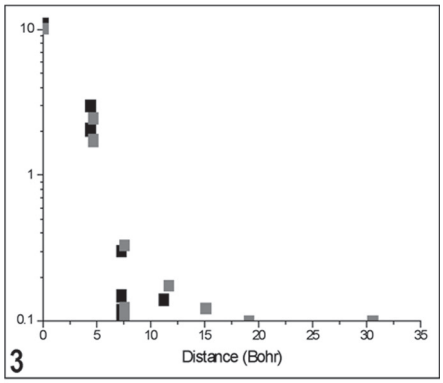

Figure 3: Force constant versus distance for 250 atom bulk silicon and bulk germanium. The black squares represent the silicon atoms and the red squares represent germanium atoms.

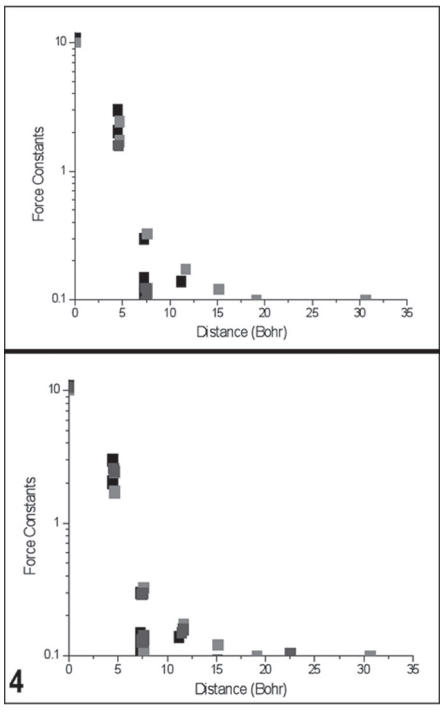

Figure 4: Force constant versus atomic distance for the 108 atom silicon-germanium interface. The black squares represent the silicon atoms, the red squares represent germanium atoms, and the blue squares represent the interface interactions. The top graph depicts an out-of-plane shift of silicon atom in the central layer while the bottom graph depicts an in-plane shift of an Si atom.