Phonon Engineering of Si/Ge Core-Shell Nanowires for Thermoelectric Applications

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INTRODUCTION

Quantum confinement in low-dimensional nanostructures is a fundamental property that increases their thermoelectric conversion efficiency, ZT [1]. Engineering the heat conduction through manipulation of phonons provides an additional route to further enhance their ZT. Parameter-free modeling approach is essential to precisely investigate how the local atomicity influences the phonon modes of the nanostructured devices. Using first principles Density Functional Theory (DFT) calculations, we have studied the structural and lattice vibrational properties of [111]-oriented Si/Ge core-shell nanowires. The detailed analysis of phonon modes shows that thermal conductance due to selective phonon modes of Si/Ge core/shell nanowires can be suppressed by engineering the ratio of core/shell atoms.

CALCULATION METHOD AND RESULTS

The calculations were performed within the framework of the first-principles density functional theory (DFT) as implemented in the Quantum Espresso Package [2]. We studied three sets of hydrogen-passivated nanowires, each has total number of atoms (excluding the hydrogen atoms) N=14 (set A), N=38 (set B), and N=74 (set C), as shown in Fig. 1. Fig. 2 shows the phonon dispersions of pristine Si and Ge nanowires in the frequency range 0-900 cm^{-1}. The highest frequency modes in the full dispersion, those originate from the vibrations of hydrogen atoms, go upto 2000 cm^{-1}, and are not shown in these plots.

The low-frequency vibrational modes of all wires show the typical one-dimensional behavior with four phonon modes. The first two modes (Fig. 3-a and b) are the flexural modes. Fig. 3-c and 3-d show the torsional and longitudinal modes. At the long-wavelength limit, flexural modes show a linear behavior, and longitudinal and torsional modes show a quadratic behavior, with respect to the phonon wave vector q.

The eigenmodes corresponding to phonon modes in the upper frequency region and lower frequency region are related to distinctly different atomic displacement patterns. The displacement of the core atoms are responsible for the eigenmodes in the lower frequency region, and the shell atom movements are more significant in the upper frequency eigenmodes. This discrepancy is visualized with two representative eigenmodes from the upper frequency region and the lower frequency region as shown in the Fig. 4.

Phonon dispersion of six core-shelll nanowires from set B are shown in the Fig. 5. A forbidden frequency region is present in all the phonon dispersions. The forbidden region divides the type of the egenmodes; core atom displacements are more responsible for the phonon modes below the forbidden regions, whereas, vibration of the shell atoms are responsible for the upper frequency region.

If we consider two nanowires with the same core atoms to shell atoms ratio, slope of the acoustic modes of the Si-shell/Ge-core nanowire is larger than that of the Si-core/Ge-shell nanowire. That suggests thermal conductivity can be suppressed by having heavier shell atoms, than having heavier core atoms. In conclusion, the detailed atomic structure plays a major role in determining the thermal conductance of the Si/Ge-core-shell nanowires.

REFERENCES

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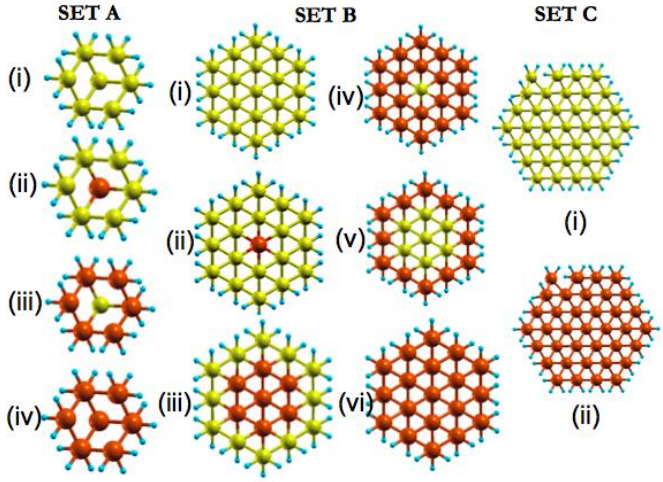

Fig. 1. Atomic configurations of all the nanowires considered in this investigation. The yellow (lighter), red (darker), and blue spheres show Si, Ge, and H atoms respectively.

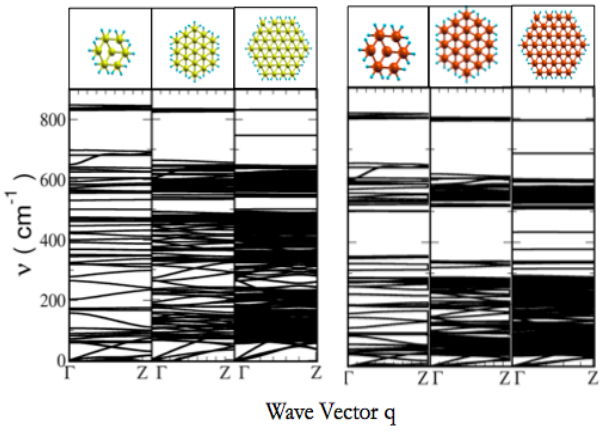

Fig 2. Lower panel shows the phonon dispersions for pristine Si (left) and Ge nanowires (right) of different diameters as shown in the upper panel.

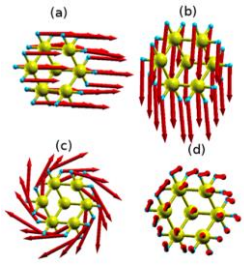

Fig. 3 Atomic displacements related to the first four lowest-frequency eigenmodes for the A-(i) Si nanowire: (a) and (b) are flexural modes (c) torsional mode and (d) longitudinal mode.

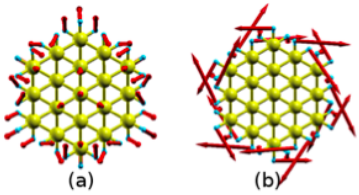

Fig. 4 Atomic displacements for sample eigenmodes for pristine set-B Si nanowires:
(a) for the lower frequency region (b) upper frequency region

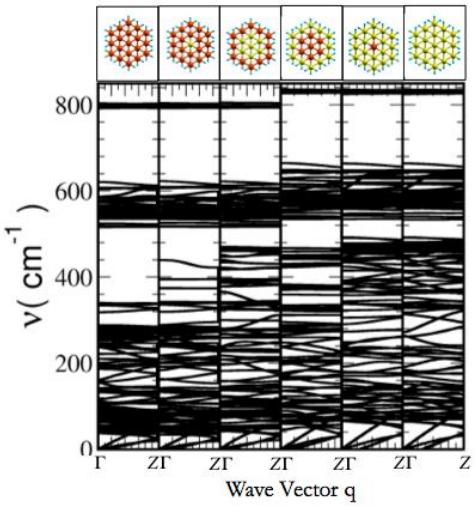

Fig 5. Phonons dispersion for all the core-shell nano wires of the set-B are shown.
The onset of each graph depicts the atomic configurations in the axial view. The left-most figure is the phonon dispersion of pristine Ge wires and the graphs are arranged with the increasing number of Si atoms.