Enhancement of thermoelectric efficiency in non-uniform semiconductor nanowires

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Abstract. Thermoelectric efficiency has been limited to a non-practical range because of the electronic material properties that are not easily controlled. Therefore, limiting thermal transport has been the best approach to enhance efficiency, and efforts to reduce the thermal conductivity even further have been extensively reported. In this work, we investigate thermoelectric transport coefficients of uniformly straight and sinusoidally undulated nanowires and compare the resulting thermoelectric figure-of-merit. The improved efficiency should suggest another strategy to effectively control thermal transport in semiconductor nanostructures.

1. Introduction

With widely growing interest in thermoelectric energy conversion, semiconductor nanowires have attracted great attention as possible candidates due to their potential for controlling both electronic and thermal transport. Recently, nanowires with diameters comparable to the phonon-mean-free-path have been studied intensively and have achieved improved thermoelectric efficiency, dictated by the dimensionless thermoelectric figure-of-merit ($ZT = S^2\sigma T/\kappa$), where S is the Seebeck coefficient, σ is the electrical conductivity, and κ is the thermal conductivity [1].

For semiconductor materials, it is very challenging to adjust the electrical power factor ($S^2\sigma$) to a desired level due to the interplay between S and σ [2]. However, the lattice thermal conductivity which has a significant contribution to the total thermal conductivity can be relatively easily manipulated by scaling, mainly owing to the phonon scattering that develops before scattering of electrons occurs in small-scale structures. The goal to limit thermal transport in ways that mildly affect electronic transport has been the most pursued strategy in thermoelectric engineering, an approach labeled as the 'phonon-glass electron-crystal' strategy [3,4].

In this study, we investigate thermoelectric properties of silicon nanowires oriented along [100] with non-uniform shape and compare their electronic and thermal transport with uniform shape nanowires, expecting an enhancement of the figure-of-merit ZT. In addition to the 1D nanowire structure in which phonons are confined in the transverse directions, non-uniformity will perturb phonon transport through the system [5]. Especially, sinusoidally undulated nanowires (sine NW) will be studied, as schematically depicted in figure 1, which can possibly realize a phonon blockade more efficiently without degrading much of the electronic transport. The length, width, undulation height, and the wavelength of the sinusoid are 108.6 nm, 4.34 nm, 4.34 nm, and 54.3 nm, respectively. The dimensions of the structure have been chosen as multiples of a lattice constant of silicon (0.543 nm) as molecular dynamics

simulation requires exact locations of atoms. The width of the wire should effectively confine both electrons and phonons in the transverse directions, while only phonons will be disturbed by the corrugated surface during transport in the longitudinal direction because of their much larger mean-free-path (a few tens of nanometers for phonons in heavily doped silicon compared to a few nanometers for electrons) [6].
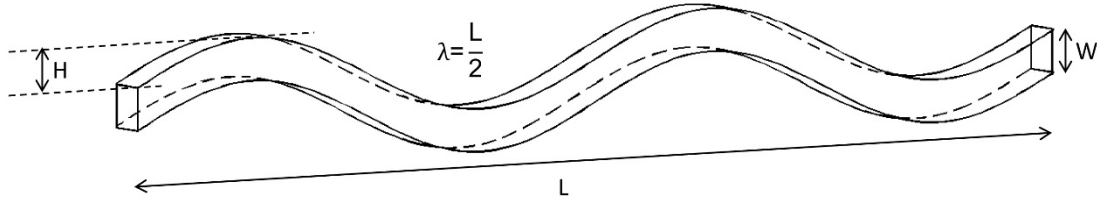

Figure 1. A schematic of a sinusoidal silicon nanowire with a square cross-section. The structural parameters are the length (L), width (W), the undulation height (H), and the wavelength of the sinusoidal wave (λ).

2. Approach

Even though the geometrical modification of nanostructures has been studied in the past, their work focused only on the degradation of thermal transport [3,7,8]. To examine trade-offs of non-uniform nanowires, studies on both electron and phonon transport should be conducted to find the maximum ZT. Loss of electronic transport due to the geometry variation can be examined by comparing an energy dependent electrical quantum conductance of the sine NWs with that of straight and uniform NWs (straight NW), employing a Recursive Green Function (RGF) approach [5,9]. The 3D conduction channel is modeled by a 1D tight-binding chain, and the quantum nature is accounted for by solving the Schrödinger equation in a series of 2D cross sections in the transverse direction. The total transmission coefficient $T(E) = \sum_{m,n}|t_{mn}|^2$ at energy E can be computed by summing the transmission probability t_{mn} over transverse sites (m,n) of each cross section [5]. $T(E)$ is then used to calculate the thermoelectric transport coefficients defined by solutions of the Boltzmann transport equation with the Landauer formalism. The electrical conductance (G) and the Seebeck coefficient (S) are expressed as [10,11]

$$G = \frac{2q^2}{h} \int_{-\infty}^{\infty} T(E)\left(-\frac{\partial f}{\partial E}\right) dE \ (1/\Omega), \tag{1}$$

$$S = -\frac{1}{qT} \frac{\int_{-\infty}^{\infty} T(E)(E-E_F)\left(-\frac{\partial f}{\partial E}\right)dE}{\int_{-\infty}^{\infty} T(E)\left(-\frac{\partial f}{\partial E}\right)dE} \ (\text{V/K}). \tag{2}$$

The thermal conductivity of the sinusoidal NWs can be computed with molecular dynamics (MD) at 300 K. We adopt the reverse non-equilibrium MD method, which imposes a heat flux first and measures the resulting temperature gradient, performed with the Stillinger-Weber interatomic potential. The MD simulation should reveal a decrease in thermal conductivity with increasing number of undulations and for larger undulation heights in the sine NWs. There must be some variants on the transport coefficients due to the strain effect if the sine NW were fabricated or bent through mechanical stress. Previous work reported enhanced electrical conductivity under bending strain [12] and reduced thermal conductivity due to uniform strain and point defects of silicon nanowires [13], which may even enhance the thermoelectric figure-of-merit. We do not consider the strain effect in this study and leave our reference structure to be oriented in [100] direction. If we were to do the actual fabrication, this structure can be realized by the photolithography process as described in [14].

3. Results

The quantum transmission of the straight and sine NWs is shown and compared in figure 2(a). The straight NW exhibits a step-function-like transmission as it represents quantum conductance at a corresponding energy level while the sinusoidal undulations directly affect the resistivity particularly at high energy. However, the undulations have little effect on low-energy transport, which leads this structure to be potentially useful of thermoelectric applications. Previous work on nanowire transport study has proven that the scattering at non-uniform surface intensifies at high energy because more conduction channels become available and efficiently couple to each other [15]. The parameters S, G, and S^2G of the wires calculated by taking into account the transmission coefficients, shown in figure 2(b-d). By definition, conductance can be converted into conductivity by multiplying L/A where A is the cross-section area. The computed S and G are within the range of experimentally measured data [3,4], but a direct comparison would not be meaningful as we consider electron scattering only due to the geometrical fluctuation and ignore otherwise. As expected, the sine NW becomes less conductive than the straight NW, reflecting the lowered transmission coefficient, and the S of the sine NW increases compared to that of the straight NW due to interplay with the conductance. Most importantly, S^2G, which is a direct measurement of the electronic effect on the ZT, decreases with the undulations above room temperature region, but the drop is not significant.
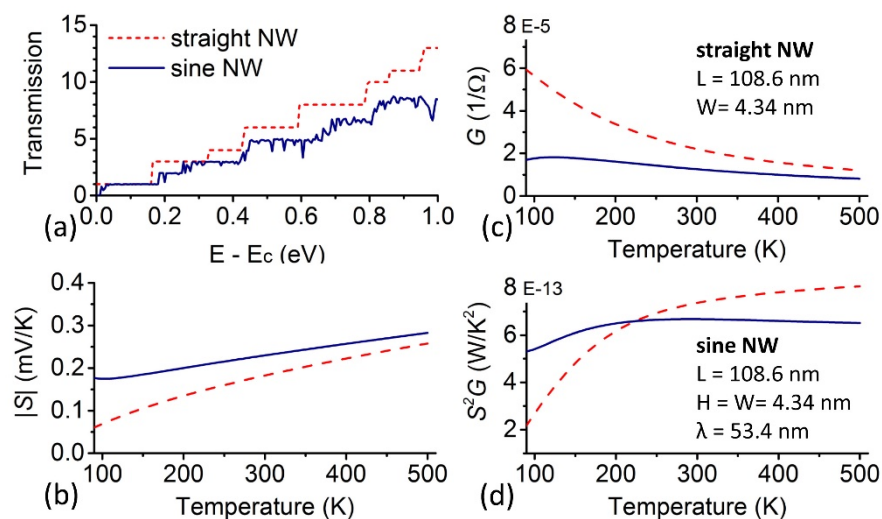

Figure 2. Comparisons of (a) electron transmission coefficient at 300 K, (b) Seebeck coefficient, (c) electrical conductance, and (d) power factor of the straight (dashed) and sine (solid line) NWs. Transmission is computed as a function of energy above the first conduction energy (E_c).

The structures under consideration have been relaxed through isothermal-isobaric and microcanonical ensemble processes for a total of 2 ns with the simulation time step of 1.0 fs, and the actual MD simulation has been performed for 2 ns. Unlike the electronic properties, κ can contribute to a great enhancement of ZT with the phonon confinement effect in low dimensional structures. This is shown in figure 3(a) where the thermal transport through the sine NW is indeed disrupted by the non-uniform geometry. Based on the calculated transport coefficients, the thermoelectric efficiency measurement ZT is computed and compared in figure 3(b) between the straight and sine NWs. The overall improvement for ZT of the sine NW is about 10 to 20 percent higher than that of the straight NW around 300 K.
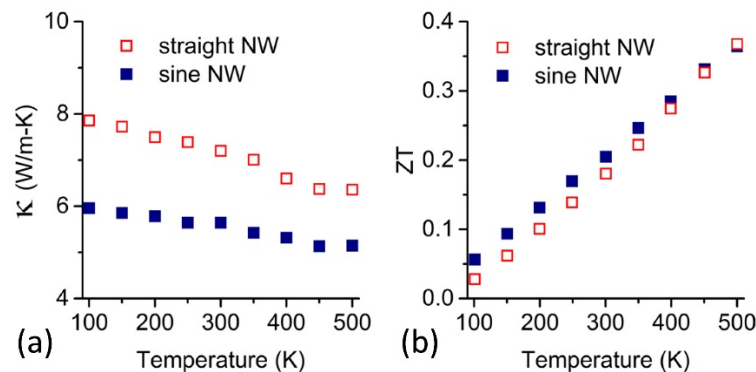
Figure 3. (a) Lattice thermal conductivity of straight and sine NWs calculated by reverse non-equilibrium MD simulation. (b) Comparison of the thermoelectric figure-of-merit ZT between straight and sine NWs.

4. Conclusion

We have investigated the thermoelectric efficiency of uniform and non-uniform shaped silicon nanowires. With the introduction of a sinusoidal-geometry variation, both electron and phonon transport are degraded, but an enhanced ZT may be achieved because there is a stronger reduction of thermal conductivity than of the electronic power factor, attributed to the large difference between electron and phonon mean-free-paths. Although the improvement obtained in this work was only around 10 to 20 percent, better enhancement can be realized by modulating structural parameters such as the cross section area, wire length, and undulation parameters. Further work will be to study benefits and downsides of the parameter modification and to find the limits of the corrugation. As we consider geometrical fluctuation as the sole perturbation to the nanowires, this strategy can be extended easily to any materials and structures.

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