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CURVATURE EFFECT ON THE THERMAL CONDUCTIVITY OF NANOWIRES

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ABSTRACT

Directional preference of the ballistic phonon transport plays an important role in the effective thermal conductivity of nanostructures. Curved nanowires can have very different thermal conductivities from straight ones. In this work, a Monte-Carlo simulator is developed and used to investigate the curvature effect on the phonon transport in silicon nanowires. The results show that the curvature of geometry does not alter the phonon transport efficiency in large wires but decreases the effective thermal conductivity in their nano-sized counterparts.

INTRODUCTION

One-dimensional materials, such as nanowires and nanotubes, received great attention over the past two decades for their potential application in thermoelectric devices and microelectronics [1]. Thermal conductivity of nanowires has been numerically calculated by many researchers including using molecular dynamics simulations [2] and the solutions of phonon Boltzmann equation [3-5]. A number of recent experiments [6,7] also suggested that that the thermal conductivity of nanowires could be one or two orders of magnitude smaller than their bulk counterparts due to both the boundary roughness and the low dimensional phonons. Past experimental and theoretical studies usually assume that nanowires are straight. However, nanowires are not always perfectly straight and the curvature of nanowires could cast uncertainty in thermal property characterization, measurement repeatability, and product reliability, due to the curvature effect on ballistic transport.

In this work, we use Monte Carlo simulation of phonon transport to study the curvature effect on phonon transport without considering the low-dimensional effect of phonons, i.e. using bulk phonon dynamic properties for the simulation. Monte Carlo technique was first used in the phonon transport study by Peterson [8] with a linear Debye theory assumption. Mazumdar and Majumdar [9] and Chen et al. [4] demonstrated size dependence of thermal conductivity in thin films and nanowires with a Monte Carlo simulator accounting for phonon polarization, nonlinear dispersion relation, and individual scattering mechanisms. Jeng et al. [10] presented a Monte Carlo simulation of Si-Ge nanocomposite and showed that the thermal conductivity of a carefully designed nanoparticle composite could be lower than the alloy limit of thermal conductivity which has significant effect on nano-thermoelectrics research. Lacroix, Joulian, and Lemonnier [11] studied the transient effect of phonon transport.

In this work, the thermal conductivity reduction in straight and curved dielectric nanowires with rectangular cross-sections are investigated and compared using the Monte Carlo simulation of phonon transport. The simulation results show that curvature plays an important role in thermal conductivity when the phonons are transported ballistically in nanowires.

THEORY

A Monte Carlo solver of phonon Boltzmann transport equation with the assumption of bulk properties are used to study the curvature effect. Here we briefly describe the methodology while interested readers are referred to Jeng et al. for details [10].

Boltzmann transport equation

Phonons are quantized lattice vibration energies and serve as the major heat carriers in dielectric materials. Under this model, particle-like properties of phonon can be introduced to replace the detailed atomic description of material [12]. The Boltzmann transport equation (BTE) describes the behavior of an ensemble of particles. With the assumptions of steady-state and absence of external force, the BTE for phonon can be written as:

$$v_g \cdot \nabla f = \left.\frac{\partial f}{\partial t}\right|_{scattering} \quad (1)$$

where $f(\mathbf{r},\mathbf{k},t)$ is the phonon distribution function and v_g stands for the group velocity of the phonons. In a detailed solution of phonon transport equation such as discrete ordinate method and Monte Carlo simulation, intrinsic scattering (RHS of Eq. (1)) is decoupled from boundary scattering while the interface and boundary conditions dictates boundary scattering.

Lattice vibrations and phonons

A gray-media approach is utilized for its computational simplicity [10], i.e., the frequency-dependent phonon properties are averaged over phonon population, and these average properties are dependent on temperature only. Since optical phonons have negligible contribution to heat transport, only acoustic phonons are taken into account [12]. This assumption is valid for most semiconductors when the group velocity of optical phonons are close to zero. In such cases, the phonon number density N is calculated as:

$$N = \sum_{p=1}^{3} \int_0^{\omega_0} \langle n \rangle \cdot D_p(\omega) \cdot d\omega \quad (2)$$

where p is index for different polarization, ω is the phonon frequency, ω_0 is the cut-off frequency which will be discussed later, $D_p(\omega) = \frac{k^2}{2\pi^2}\frac{\partial k}{\partial \omega}$ is the phonon density of state for each branch, $\langle n \rangle = \left[\exp(\hbar\omega / k_B T) - 1\right]^{-1}$ is the equilibrium phonon distribution function, $k_B = 1.38 \cdot 10^{-23} J/k$ is the Boltzmann constant, and $\hbar = 1.05 \cdot 10^{-34} J \cdot s$ is the reduced Plank constant. In this work, the study is conducted for silicon nanowires by assuming phonon dispersion in silicon nanowires will not be altered from bulk dispersion. Following reference [15], a sine-type approximation $\omega = \omega_0 \sin(\pi k / 2k_0)$ is used for the dispersion relation of silicon acoustic phonons, where ω_0 is the cut-off frequency and k_0 is the cut-off wave vector. Detailed calculation of ω_0 and k_0 can be found in the appendix B of reference [15].

The internal energy density E is the total energy of phonon under a certain temperature:

$$E = \sum_{p=1}^{3} \int_0^{\omega_0} \hbar\omega_p \cdot \langle n \rangle \cdot D_p(\omega) \cdot d\omega \quad (3)$$

and the average phonon frequency ω_{avg} is defined as the internal energy density divided by phonon number density and the reduced Plank constant:

$$\omega_{avg} = \frac{1}{\hbar \cdot N} \sum_{p=1}^{3} \int_0^{\omega_0} \hbar\omega_p \cdot \langle n \rangle \cdot D_p(\omega) \cdot d\omega \quad (4)$$

The heat capacity of acoustic phonons $C_{acoustic}$ is written as

$$C_{acoustic} = \frac{\partial E}{\partial T} = \sum_{p=1}^{3} \int_0^{\omega_0} \hbar\omega_p \cdot \frac{\partial \langle n \rangle}{\partial T} \cdot D_p(\omega) \cdot d\omega \quad (5)$$

and the group velocity v_{avg} is calculated as:

$$v_{avg} = \frac{1}{N} \sum_{p=1}^{3} \int_0^{\omega_0} \frac{\partial \omega_p}{\partial k} \cdot \langle n \rangle \cdot D_p(\omega) \cdot d\omega \quad (6)$$

A lumped phonon mean free path (MFP) is found via the simple kinetic theory as Eq. (7), where λ_{bulk} is the MFP of bulk material and k_{bulk} is the measured thermal conductivity of bulk material [13,14]:

$$\lambda_{bulk} = \frac{3 \cdot k_{bulk}}{v_{avg} \cdot C_{acoustic}} \quad (7)$$

Monte Carlo simulation

For simplicity of calculation, we assume nanowires with square cross-sections rather than a cylindrical one. Figure 1(a) and 1(b) show two geometries under consideration, which correspond to straight and curved wires respectively while the heat is applied in the length direction along the wires. The simulation domain is divided into many "subcells", as seen in Fig. 1(a) and 1(b). At the end of each time step, phonon energy within each subcell is calculated and converted into the effective local temperature and thus the local phonon MFP can be calculated.

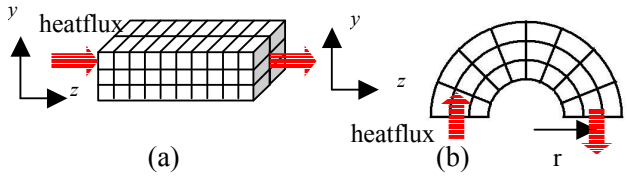


Figure 1. (a) Rectangular domain, (b) Curved domain (square cross-section of curved wires is not shown in the figure).

In a Monte Carlo simulator, phonons are tracked as they drift and collide within the computational domain [10]. They encounter various scattering events including boundary and intrinsic scatterings while they drift. The probability for a phonon to experience intrinsic scattering during a time period dt is given by $P_s = 1 - \exp(-v_{avg} \cdot dt / \lambda_{bulk})$, where the phonon mean free path λ_{bulk} is calculated using Eq. (7). For each phonon, a random number is drawn and compared with P_s. If the random number is less than P_s, then the phonon will be scattered and reassigned a random flight direction. The energy of the scattered phonon is then set to comply with the local subcell temperature in order to establish a local equilibrium between the phonon and the subcell. Energy surplus/deficiency due to intrinsic scattering is monitored at the subcell level. When the accumulated energy difference exceeds the phonon unit energy, a phonon is generated or deleted. During boundary interactions, the phonon is either specularly or diffusively reflected. Only the flight direction is altered in this process. Surface roughness is

described by the fraction of specularity p between 0 and 1, where p=0 represents diffusive reflection and p=1 represents specular reflection.

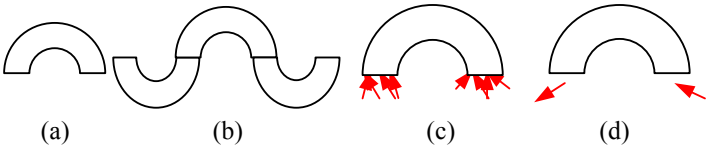
Heat flux is prescribed to a fixed value in the heat flow direction. A certain number of phonons are emitted into the computational domain, and the number is calculated so as to ensure that the net heat flows across the two ends are identical and equal to the prescribed value. Thermal properties of emitted phonons are determined by extrapolating the internal temperature profile to the boundaries. Detailed description to heat flux treatment can be found in [10].

Simulations are conducted for Si nanowires at 300K. The phonon MFP at this temperature is about 220nm. Time step is chosen to achieve half subcell length per stride, and the phonon bundle number is controlled around 50 per subcell.

RESULTS AND DISCUSSION

Length effect on thermal conductivity

We investigated the thermal conductivity of a single curved wire segment (half circle) and an infinitely-long periodic curvy wire (which comprises many half circles periodically) as illustrated in Figs. 2(a) and 2 (b) while the corresponding boundary conditions for numerical simulation are shown in Figs. 2(c)-(d). For a wire segment, phonon blackbody emission boundary condition is used for which phonons are injected into computational domain with random azimuthal direction. For an infinitely-long periodic curvy wire, the periodicity in boundary condition is realized by assigning the flight direction, position, and remaining flight time of a phonon leaving the computational domain to a phonon entering the domain from the opposite side. In this study, nanowire cross section is 50nm by 50 nm with various curvatures. Curvature is the inverse of the radius of the half circle and is calculated as π/half circumference. Fraction of specularity p is set to zero, which assumes a totally diffusive surface. Fig. 3 shows that the thermal conductivity obtained from emission boundary case is always lower than the periodic boundary case. It is because in periodic boundary condition, phonon direction is biased toward the already-imposed heat flux. In the emission boundary condition, emitted phonons have hemispherically uniform distributed flight directions, and the resulting heat transport is not as efficient as in the periodic case. This length effect also diminishes as wire curvature decreases, as shown in Fig. 3.



(a) (b) (c) (d)

Figure 2. (a) Curved wire segment (b) Periodically long curvy wire and their corresponding boundary conditions in numerical simulations: (c) Emission boundary condition (d) Periodic boundary condition

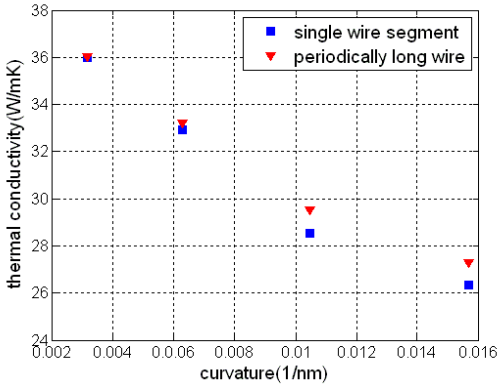


Figure 3. Thermal conductivity of Si nanowires with 50nm by 50nm cross section at 300K.

Effect of wire thickness on thermal conductivity

Decreasing wire thickness increases the surface-to-volume ratio of the wires. This would reduce the effective thermal conductivity of wires due to boundary scattering, especially when the boundaries are highly diffusive with large p. Curvature effect is evaluated by dividing the thermal conductivity of a periodically long curved wire (curvature as $0.0157nm^{-1}$) with the thermal conductivity of a straight wire (curvature as zero). The wire thickness is 20nm, 30nm, 40nm, and 50nm respectively. Fig. 4 shows the thermal conductivity ratio of periodically curved nanowires to straight ones with the same cross-sectional areas, and it is seen that decrease of fraction of specularity or decrease of wire thickness results in a higher reduction of thermal conductivity. For example, the thermal conductivity of a curved nanowire with 20nm by 20nm cross area and $0.0157nm^{-1}$ curvature is only 65% of its straight counterpart. The phonon transport in a curved nanowire is impeded by the multiple collisions with the boundaries, while this boundary collision is less seen in the straight case.

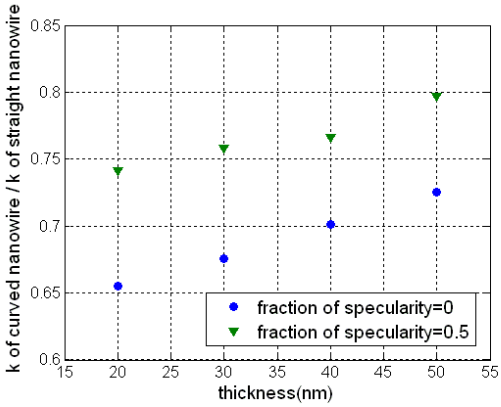


Figure 4. Thermal conductivity ratio of curved and straight Si nanowires at 300K

Curvature effect on thermal conductivity

Curvature effect is studied by comparing the thermal conductivity of a periodically curved nanowire and straight one

under different curvatures. The cross section of the wire is kept constant at 50nm by 50nm, and the curvatures are 0.00314nm^{-1}, 0.00393nm^{-1}, 0.00524nm^{-1}, 0.00785nm^{-1}, and 0.0157nm^{-1}, which correspond to periodic arc length of 1000nm, 800nm, 600nm, 400nm, and 200nm, respectively. We assume purely diffusive (p=0) in our calculations. Fig. 5 shows the thermal conductivity ratio of periodically curved nanowires to straight ones with the same cross-sectional areas. A reduction in thermal conductivity by increasing the curvature is clear seen in Fig. 5. A curved nanowire with curvature of 0.0157nm^{-1} can have only 72% of its straight counterpart. Since ballistic phonons rarely experience intrinsic scattering, boundary scattering dominates in the scattering process. Due to the irregular geometry of curved nanowires, ballistic phonons experience more boundary scattering in a nanowire with larger curvature. This in turn retards phonon transport and reduces the effective thermal conductivity.

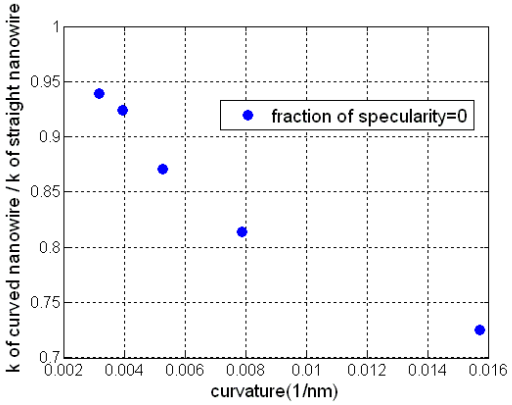


Figure 5. Thermal conductivity ratio of Si nanowire with 50nm by 50nm cross section and different curvatures at 300K

CONCLUSION

This work presents a study on the curvature effect on the thermal conductivity of nanowires using the Monte Carlo simulations of phonon Boltzmann equation assuming bulk phonon properties (density of states and group velocity). Straight and curved nanowires with rectangular cross-sections are investigated and compared. The simulation results show that the curvature plays an important role in thermal conductivity when the phonons are transported ballistically in nanowires.

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