International Journal of Heat and Mass Transfer 114 (2017) 550–558

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Heat transport in phononic-like membranes: Modeling and comparison with modulated nano-wires

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ARTICLE INFO

Article history:
Received 24 April 2017
Received in revised form 14 June 2017
Accepted 15 June 2017

Keywords:
Nano-structuration
Silicon
Molecular Dynamics
Monte Carlo
Thermal conductivity

ABSTRACT

The thermal conductivity of phononic-like membranes and their analogous nano-wires are computed with both Molecular Dynamics and Monte Carlo simulations. Using both methods, the thermal conductivity of the porous membranes with cylindrical pores is found to be lower than the nano-wire's one for the same scattering surface to volume. This work confirms experimental observations by Nomura et al. (2015) and Yu et al. (2010) for similar systems. The lower thermal conductivity of membranes is attributed to the different orientation of the scattering surfaces either at the pores or at free surfaces and not to coherent effects. The comparison between the porous membranes and nano-wires with periodic cylindrical modulations shows that thermal transport is hindered further when there are surfaces perpendicular to the heat flow. These results show that coherent effects have no impact on thermal transport in both kind of nano-structures at room temperature.

1. Introduction

During the last decades, increased interest has been observed to tailor different physical properties of nano-structures and nano-structured materials. Unique characteristics and effects are found in nano-structured semi-conductors compared to their bulk properties. Silicon is the most common semi-conductor on Earth and is a non-pollutant material. Therefore, nano-structured Silicon has been intensively studied experimentally and theoretically for its optical [3,4] or thermal properties [5–7] among a plethora of other properties. The importance of thermal management of nano-structures has arisen as the electronic devices became smaller and more powerful. Nano-structured semi-conductors are widely used for energy thermoelectric conversion [8,9] or in photovoltaic devices [10].

Efficiency of thermoelectric materials depends on their electrical to thermal conductivities ratio (figure of merit), thus the thermal conductivity (TC) has to be very low. However, current thermoelectric materials have a poor figure of merit and cannot be massively used for energy production. Then, the aim of the nano-structuration of semi-conductors is often to reduce the thermal transport by multilength scale hierarchical structuring with purpose to increase the density of interface or introducing point or extended defects. On the other hand as electrons have a much lower mean free path than phonons [11], it can be assumed that their conductivity is preserved in most of the nano-structured semi-conductors.

Historically, the study of heat transfer in composite materials began with Maxwell [12] and Lord Rayleigh [13] at the end of the 19th century. Thereafter, thermal transport in nano-structured compounds gained interest, including porous and nano-porous materials [14–16]. Different kinds of nano-structuration are possible today, especially with the fast development of fabrication methods. The TC of super-lattices alternating layers of different semi-conductors have been intensively investigated during the last decade [17–19]. In such structures, band folding plays a key role in TC lowering. The reduction of the thermal transport was also observed in thin films [20–23] and nano-wires [24–29]. In the recent years, several studies are devoted to thermal behavior of bulk nano-porous systems [30–33], or nano-porous membranes [34–38].

Recently, a new method has been proposed to reduce thermal transport in periodic nano-structures. By analogy to the photonic crystals, which are also known as photonic band gap materials as there are forbidden photon propagation frequencies [39,40], an artificially and periodically structured material could remove phonons of certain frequencies [41,36,42–44]. These phonon band gaps

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http://dx.doi.org/10.1016/j.ijheatmasstransfer.2017.06.067
0017-9310/

are due to scattering and interference phenomena, the latter being known as coherent effects. If the prohibited phonon modes are heat carriers, the TC would be further reduced [45]. Abnormally low TCs have been experimentally observed in periodically nanostructured Silicon [2] and this was attributed to these coherent effects.

Nomura et al. compared the experimental TCs of membranes with periodic cylindrical pores to those of nano-wires [1]. They found that thermal transport is weaker in membranes, despite their low scattering surface to volume ratio compared to nanowires with similar characteristic size. This might be due to phononic effects (i.e. any effect related to a modification of phonon dispersion properties like band flattening, band gap . . .or coherent scattering of phonon due to the periodicity of the hole pattern), but authors mentioned that the "local angles between local and global heat flux" could be responsible of this phenomenon [1]. In other words, they point out the different impact of scattering surfaces on thermal transport according to their spatial orientation compared to the direction of the main thermal gradient. At this stage a simple schematic description of phonon transport can be depicted. When the TC is measured along the growth direction of a nano-wire, all scattering surfaces of the system are parallel to the heat flux. If phonons are diffusely scattered at the free surfaces, half of them are back-scattered (Diffuse Mismatch Model). In contrast, in the "phononic crystal" (hollow membrane) the surface of the pore (which is here a part of the scattering surfaces with upper and lower boundaries) is mainly perpendicular to the heat flux (see Fig. 1a). Then, the back-scattering of phonons in the membrane is increased as compared to the nano-wire and the TC of the membrane is reduced.

In this work, the same strategy as Nomura et al. has been exploited, making the comparison between porous membranes and nano-wires with two numerical techniques: Monte Carlo solving of the BTE and Molecular Dynamics. The comparison between the two nano-structured materials shows the importance of the orientation of the scattering surfaces on thermal transport. Technical details about the computations of the TC with these two methods are presented in Section 2, then the geometries of the modeled systems are defined in Section 3. TC experimental results and numerical simulation results are compared in Section 4. In the same section, the phonon densities of states computed with Molecular Dynamics are presented to give physical insights. Finally, in Section 5 modulated nano-wires are modeled and their TC is given in comparison to membrane's TC with the same geometric parameters.

2. Simulation methods

2.1. Molecular Dynamics

Molecular Dynamics simulations are performed with *LAMMPS* [46]. Equilibrium Molecular Dynamics (EMD) method is used to determine the TC via the Green-Kubo formula [47]:

$$k_{xy} = \frac{\mathcal{V}}{k_B T^2} \int_0^{+\infty} \langle J_x(t) J_y(0) \rangle dt \quad (1)$$

where x and y subscripts denote space directions, $\mathcal{V}$ is the system's volume, J is the heat flux, t is the time, T is the system's temperature and k_B the Boltzmann constant. Then, the TC can be obtained for all directions. For membranes, the flux auto-correlation function is calculated in x and y directions ($J_x(t)J_x(0)$ and $J_y(t)J_y(0)$, respectively). The in-plane TC is the average of the two thermal conductivities in direction x and y (see Fig. 1a). For nano-wires, the flux auto-correlation function is calculated only for x direction which is the growth direction of nano-wires (see Fig. 1b). The interactions between the atoms are described with the Stillinger-Weber potential [48] with modified coefficients by Vink et al. [49] as this latest parameterization is able to describe thermal transport in Silicon, in both crystalline and amorphous phases.

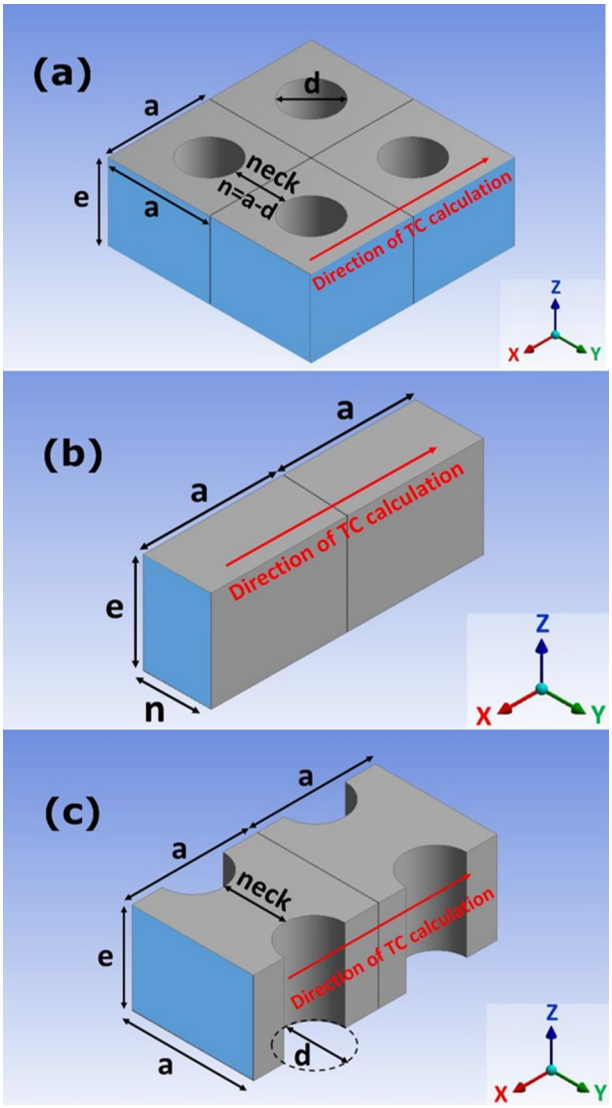

Fig. 1. Schematic representation of the modeled systems. (a) Phononic-like nanoporous membranes (np-memb). Blue faces indicate periodic boundary conditions in these directions, the other direction is not periodic as it is the direction of the thickness of the membrane. (b) Equivalent nano-wire (NW) whose width is equal to the neck between two adjacent pores in the porous membrane. The height is equal to the thickness of the membrane. The blue face indicates a periodic boundary condition in this direction (length of the nano-wire), the other directions are not periodic (walls of the nano-wire). (c) Equivalent modulated nano-wire (m-NW). Its width is equal to the periodicity a of the porous membrane and is modulated by periodic and cylindrical constrictions with the same diameter d as the pores. Thus, the minimum width is equal to the neck between two neighbours pores in the membrane. The blue face indicates a periodic boundary condition in this direction (length of the m-NW), the other directions are not periodic (walls of the m-NW). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The time step is set to 0.5 fs in all EMD runs. At the beginning of the simulation, the system is relaxed at $T = 300$ K with a NVT ensemble during 200 ps. Then the computation of the TC is performed every 40 ps under a NVT ensemble, estimating the flux fluctuations correlation every 10 fs, for a total duration of 10 ns. The TC is averaged over the second half of the simulation, when