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A comparative study of molecular dynamics simulation methods for evaluation of the thermal conductivity and phonon transport in Si nanowires

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ABSTRACT

In the present study, various MD methods including Equilibrium Molecular Dynamics (EMD) and two different Non- Equilibrium Molecular Dynamics (NEMD) methods are studied and compared for evaluation of the thermal conductivity and total and partial phonon density of states spectra in silicon nanowires, as a case study. The thermal conductivity of nanowires was determined as a function of length, cross section width and temperature. According to the results obtained via various MD methods, the thermal conductivity increases by increasing the length and cross section or decreasing the MD temperature. However, it was observed that despite the same initial conditions, different MD methods could predict considerably different values for the thermal conductivity which was found to be due to the different equilibrium temperature achieved in different methods. The total phonon density of states spectra was then employed to analyze the phonon transport properties of a 45 nm SiNW simulated using various MD techniques. Two major peaks were observed at around 16.5 and 5 THz which are attributed to Si-Si bond modes. Finally partial phonon density of states was calculated to differentiate the contributions to the phonon DOS from surface atoms compared to central atoms.

1. Introduction

Thermal properties of nanostructure materials have recently received extensive attentions due to their importance in a variety of high tech applications, e.g. the design of nanoscale thermoelectric energy generators, on-chip coolers, nanoelectronics and energy conversion devices [1–4]. Silicon nanowires (SiNW) is our current interest due to its applications in areas ranging from field effect transistors, interconnects and nanoelectronics to thermoelectrics resulted by their remarkable electronic and optical properties as well as their compatibility with traditional Si-based electronic devices [3,5,6]. Thermal properties of SiNWs have been experimentally investigated in a number of works. Li et al. [2] measured the thermal conductivity of silicon nanowires as a function of diameter and temperature ranging from 22 to 115 nm and 20 to 320 K, respectively. Phonon boundary scattering and phonon spectrum modification were reported as the major reasons for the strong dependence of thermal conductivity to diameter. Pan et al. [4] fabricated vertically aligned SiNWs and germanium doped SiNWs with 100 nm diameter. They showed that surface doping of Ge on silicon nanowire reduces the thermal conductivity by 23% at room temperature. Glassbrenner et al. [7] measured the thermal conductivity of single crystal silicon from 3 to 1580 K. They found that the electronic contribution to the thermal conductivity appears at temperatures above 1000 K.

Experimentally determination of the thermal properties of nanowires is difficult and sometimes impossible. Molecular dynamics (MD) simulation has been widely used to study the thermal properties of nanowires, as an alternative robust and reliable method.

In general, three MD methods are available for studying the thermal conductivity of nanostructures. The first method is Equilibrium Molecular Dynamics (EMD) which is based on the Green-Kubo relation. For example, Ramazani et al. [8] calculated the thermal conductivities of α, β and γ graphene nanotubes (GNTs) as well as of carbon nanotubes (CNTS) using Green-Kubo formula in a wide range of temperature from 50 to 400 K. Li et al. [9] studied the phonon thermal conductivity of hydrogenated silicon nanowires using EMD. Their results revealed that the surface hydrogen passivation increases the thermal conductivity in comparison with pure SiNWs. Donadio et al. [10] applied EMD to

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study the lattice thermal conductivity (κ) of silicon nanowires as a function of temperature. They showed that in wires with amorphous surfaces, κ may reach values close to that of the amorphous silicon and is nearly constant between 200 and 600 K which is due to the presence of a majority of non-propagating vibrational modes. They also showed that the presence of a thin amorphous surface layer may reduce the thermal conductivity of thin SiNW by up to 100 times with respect to the bulk value. Sebastian et al. [11] employed EMD simulations to measure the thermal conductivity of silicon nanowires in various temperatures and cross-sections for both rigid and free boundary conditions. They found that phonon group velocity and the scattering mechanism may also be responsible for the thermal conductivity reduction.

The other MD methods which are applied to study the thermal transport properties of nanowires are Non-Equilibrium Molecular Dynamics (NEMD) and Reverse Non-Equilibrium Molecular Dynamics (RNEMD) which are based on the Fourier's law of heat conduction. Wang et al. [12] used NEMD to study the dependence of the thermal conductivity of silicon nanowires on the wire length, cross-sectional area, and temperature. They showed that the nanowire length dramatically reduces the effective phonon mean free path in the nanowire, which reduces its thermal conductivity. Abs da Cruz et al. [13] applied RNEMD to study the influence of various interatomic potentials in predicted thermal conductivity of Silicon nanowires. They showed that the best interatomic potential is the second nearest-neighbor modified embedded atom method potential followed by the Stillinger-Weber, and then the Tersoff III. They also found that as the simulated nanowires are perfect and without surface oxide or surface roughness, the thermal conductivities are overestimated compared to real nanowires. Huang et al. [14]. Performed NEMD simulations to investigate the thermal transport in Si/Ge nanocomposites. They found that the thermal conductivity of the nanocomposite seems to have weak dependence on the bulk temperature (200–900 K). Termentzidis et al. [15] predicted the thermal conductivity of polytype modulated SiC nanowires using NEMD and reported that the thermal conductivity of the diameter modulated nanowires may be even smaller than that of the constant diameter nanowire with the small section. This remarkable reduction in thermal conduction was attributed to a significant thermal boundary resistance displayed by the constriction, as measured by independent molecular-dynamics simulations.

The aim of the present study is to introduce the EMD and two different NEMD methods and compare their performance in evaluation of the thermal properties of SiNWs. Various SiNWs with different lengths, cross section widths and equilibrium temperatures are studied. More ever, a quantum correction is implemented for the MD predicted thermal conductivity to take into account the quantum effects, especially for temperatures below the Debye temperature. For more detailed analysis, the total and partial vibrational density of states acquired from MD simulations are exploited to study the effect of temperature on density of state (DOS) and to distinguish the contributions of silicon atoms in the surface and central areas of silicon nanowire.

2. Theory and methods

2.1. Simulation details

The thermal transport properties of silicon nanowires with lengths of 20 nm, 30 nm, 45 nm and 60 nm and cross-section widths of 1.2, 2.2, 3.3 and 4.4 nm were studied using EMD and two various NEMD simulations. Furthermore, for the nanowire with 20 nm length and 2.2 nm cross section width various operational conditions which can result in different equilibrium temperature were studied. In all simulations, the Stillinger-weber (SW) potential was employed to model the interactions between silicon atoms. This potential has two and three body interaction terms to model the faces in the diamond lattice [16]. To avoid any anomalous phenomena such as nanowire deformation, bending, twisting, etc, fixed potential walls were placed around the nanowire parallel to the longitudinal z direction, as proposed by Cruz et al. [13]. The fixed potential walls were created by placing silicon atoms at a fixed distance R from the silicon nanowire surfaces. Using the Lennard-Jones (9/3) potential stated as:

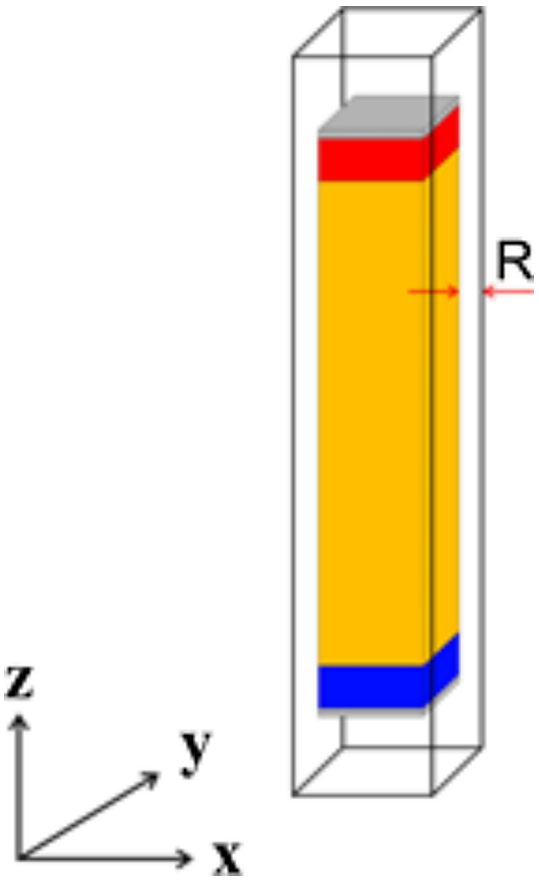

Fig. 1. Heat and cold reservoirs in addition to the potential walls parallel to the longitudinal direction of the silicon nanowire.

$$E = \varepsilon\left[\frac{12}{5}\left(\frac{\sigma}{R}\right)^{9} - \left(\frac{\sigma}{R}\right)^{3}\right], \tag{1}$$

where ε and σ are the Lenard-Jones parameters and R is the distance from the particle, the walls were located at $R = 1.2009\sigma$ from nanowire surface. At this distance, the attraction and repulsion forces on the nanowire are equal. To avoid any undesired movement of the nanowire during the simulation, the atoms in the first and last 1 nm of the nanowire in the longitudinal direction were frozen. More ever, the two 4 nm slabs shown in Fig. 1 with red[1] and blue colors were assigned as heat source and sink, respectively, and are called hot and cold reservoirs hereafter. The simulations were performed using Large Scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package [17]. The velocity Verlet algorithm with the time step of 0.5 fs was applied to solve the equations of motion. To obtain the initial configuration the potential energy was minimized using the conjugate gradient method. The nanowire was then equilibrated at the constant temperature of 300 K in a canonical ensemble (NVT) using the Nose-Hoover thermostat for

[1] For interpretation of color in Fig. 1, the reader is referred to the web version of this article.