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Investigation of thermal rectification in bi-layer nanofilm by molecular dynamics

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ABSTRACT

The rectification of the cross-plane thermal conductivity and the interfacial thermal resistance of nanoscale bi-layer films are investigated using the nonequilibrium molecular dynamics method with two different heat baths. The effects of the thickness of the single layer with the total film thickness fixed, the atomic mass ratio and temperature difference across the film on the thermal rectification are considered. The results show that the thermal rectification is dependent on the temperature gradient and the atomic mass ratio. The thermal conductivity is usually larger when the heat flux direction is from the light mass layer to the heavy mass one. It is found, however, that the rectification could be reversed if the heavy layer is thin enough. The phonon density of states is given to explain the mechanism behind the phenomena. The overlap of the phonon density of states of the two layers is almost same even if the rectification of the thermal conductivity is reversed.

1. Introduction

The researches on the nanoscale heat conduction have been enriching our knowledge on the nanoscale in the past decades, such as the size effect on thermal conductivity, the thermal resistance at interfaces between solids or the grain boundaries. One of the novel findings, the thermal rectification in nanoscale heat conduction [1–3] gradually attracts more and more attentions in recent years. The thermal rectification means that the thermal conductivity or conductance is different with opposite heat flux directions. Heat can run preferentially in one direction but not in the opposite direction with the same temperature difference. Its electrical counter device in extreme case is the diode that is among the most crucial achievements among the modern technologies.

In 2002, there were two reports on the thermal rectification, one was the one-dimensional nonlinear lattice [3] and the other was the heat conduction in the radial direction of argon nanotubes [2]. Since then, many theoretical models of thermal rectification [4–9] were proposed based on the nonlinear lattices that include one-dimensional, two-dimensional, two-segment and triple-segment nonlinear lattices. In 2006, Chang et al. [1] reported an experiment on nanoscale solid-state thermal rectifier by using unevenly mass-loaded carbon and boron nitride nanotubes, which was an attempt toward the experimental demonstration of the thermal rectifier.

In recent years, the thermal rectification was predicted in one-dimensional lattices with mass gradient [10] and many carbon nanostructures such as carbon nanotube intramolecular junctions [11], carbon nanohorns [12], carbon nanocone [13], graphene nanoribbons [14,15]. In these nanostructures, one common feature is that the heat flux runs preferentially along the direction of decreasing characteristic size and the thermal rectification is strongly dependent on the temperature gradient. The mechanism of the thermal rectification was attributed to the better overlap of the phonon spectra of the two particles around the connecting parts when the direction of temperature gradient is from the larger characteristic size to the smaller one.

The thermal rectification properties were also observed at the solid–solid interfaces [16–18] and solid–liquid interface [19]. Due to the different lattices between two real crystal materials, it is hard to construct a solid–solid interface between two solid crystals with periodic boundary condition in molecular dynamics (MD) simulation. Liang [16] simply changed the atomic mass in different layers and investigated the thermal rectification in the normal conduction of a bi-layered nanofilm with solid argon structure by nonequilibrium molecular dynamics (NEMD) method. The average temperatures with opposite heat flux directions are different because of the constant heat flux used, which is possibly one of the causes of thermal rectification. The investigation on the thermal rectification of the bi-layered nanofilm by NEMD [17] demonstrates that the

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Nomenclature

D	Phonon density of states
J	Heat flux density in the nanofilm
L	Thickness of bi-layer nanofilm
L_A	Thickness of Layer A
L_B	Thickness of Layer B
R	Rectification efficiency of thermal conductivity
S	Cross-sectional area of the nanofilm
T	Average temperature
T_A	Temperature of the heat bath in the layer A
T_B	Temperature of the heat bath in the layer B
ΔT	Half of the temperature difference across the film
δT	Interfacial temperature difference
κ	Thermal conductivity
ω	Angular frequency of phonon
γ	Atomic mass ratio of atoms in Layer B and Layer A

rectification of the interfacial thermal resistance decreases with increasing temperature, which cannot be interpreted only by the difference of the interfacial temperature. Hu et al. [18] made a realistic interface model system composed of crystal silicon and amorphous polymer and predicted the thermal rectification. Hu et al. [19] further constructed a solid–liquid interface model consisting of silica, self-assembled monolayers and water. Their result demonstrates that the thermal rectification is up to 54% with a heat flux of 2700 MW/m^2.

Many factors could affect the thermal rectification as shown in the reports mentioned above, such as inter-atomic potentials, temperature and temperature gradient, materials, structure, variation in size, interface, etc. Many researchers attributed the mechanism to the overlap of phonon spectra [10–15], but Hu et al. [20] found this explanation is not true for strong interfacial interaction because of the band mixing of the segments. In fact, the interfacial resistance cannot be explained solely in terms of the mismatch of phonon spectra [21]. The investigation of bi-layer nanofilms by Ju et al. [22] demonstrated that a larger overlap of phonon spectra leads to a smaller interfacial resistance, but the overlap explanation is not quite reasonable for the rectification. They observed a change in interfacial thermal resistance about 15% with opposite heat fluxes although the overlap of phonon density of state (DOS) is almost unvaried, and found that the relationship between the interfacial thermal resistance and overlap DOS area is not monotonic.

This paper reports the investigation of the thermal rectification of bi-layer nanofilms by NEMD method. The focus is on the effects of layer thickness and atomic mass, as well as temperature gradient. The mechanism of the rectification is discussed.

2. Simulation model

The bi-layer film is composed of two dielectric thin layers, Layer A and Layer B. Layer A is solid argon and Layer B is the same as Layer A except for its atomic mass. The atomic mass of Layer B is a multiple of argon atom mass. For convenience, the atomic mass ratio of Layer B and Layer A is denoted as γ. Each layer is divided into the inner heat conduction zone, the heat bath zone, and the outmost fixed wall zone as shown in Fig. 1. It is necessary to define a basic system thickness because the thermal conductivity depends on the system length when the mean free path (MFP) of phonon is equivalent to the layer thickness. Let L_A and L_B represent the thicknesses of the heat conduction zones of Layer A and Layer B, respectively. The

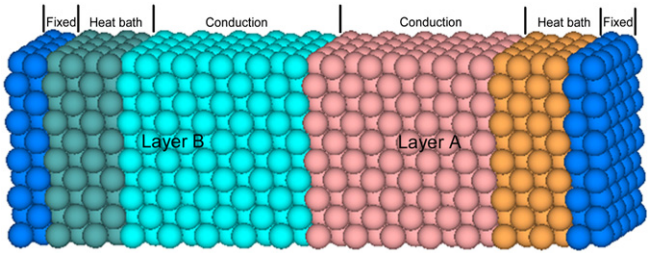

Fig. 1. A typical bi-layer film structure composed of two materials. The thickness of Layer A and Layer B is 5 UCs, and the cross-sectional area is 5 UCs × 5 UCs. The regions marked with "Fixed" are fixed in the MD process, and the regions marked with "Heat bath" are the heat bath zones.

system thickness is defined as $L = L_A + L_B$. Fig. 1 demonstrates a typical bi-layer film structure in which L_A and L_B are both five unit cells (UC) and the cross-sectional area S is 5 UCs × 5 UCs. The thickness of one UC contains two atomic planes for the face-centered cubic of solid argon. The cross-sectional area is large enough to eliminate the boundary effect on the MD simulation of the normal thermal conductivity of films [23]. The outmost fixed wall zones (marked as region "Fixed" in Fig. 1) with one UC thickness mean that the atoms in the zones are fixed in the simulations. The heat bath zones (marked as region "Heat bath" in Fig. 1) are both taken two UCs thick to assure that the atoms in heat conduction zones cannot interact with those in the fixed wall zones. Constant temperature in each heat bath is realized by two methods. The first is to rescale the velocities of the atoms in the zone with the total net momentum at constant [24]. The heat flux through the conduction zone can be calculated by the change in kinetic energy due to the velocity rescale. The second is realized by the generalized Langevin equation (GLEQ) approach [25,26] in which two additional forces, a drag force being proportional to the atom velocity and a random force, are added into the Newtonian equation of the motion. The way of obtaining the heat flux is the same. The simulation results from these two methods are compared. The temperatures of the heat baths neighboring the conduction zones of Layer A and Layer B are denoted as T_A and T_B, respectively. For convenience, two quantities are introduced, half of the temperature difference across the conduction zones $\Delta T = (T_A - T_B)/2$, and the average temperature $T = (T_A + T_B)/2$.

All the atoms in the conduction and heat bath zones interact with each other through the classical Lennard-Jones (12–6) potentials [27],

$$\phi(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right],$$

where r is the inter-atomic distance, ε is the energy parameter, and σ is the atom diameter. The potential parameters ε and σ are taken from the argon crystal, which are 1.67×10^{-21} J and 3.40×10^{-10} m, respectively [27]. The periodic boundary condition is applied in the in-plane directions that are normal to the heat flux direction (x axis). The velocity Verlet method is employed to integrate the equations of motion with a time step of 4.3 fs. In the MD simulation, calculating the inter-atomic potential and force consumes most of the whole simulation time. To reduce the calculation, a potential cutoff radius of $r_{cut} = 2.8\sigma$ and a neighbour list method with a skin thickness 0.4σ are used [27]. The simulations last for 1×10^6 or 4×10^6 time steps (for the film thickness of 10 UCs or 40 UCs), which are 4.3 or 17.2 ns. The first half of simulation steps is used to equilibrate the system and to relax the initial stress and the second half is used to make the statistic average of the heat flux. The effective cross-plane thermal conductivity κ can be obtained according to the Fourier's law,