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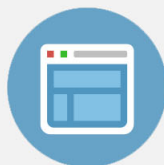
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Molecular dynamic simulation of diamond/silicon interfacial thermal conductance

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Non-equilibrium molecular dynamic simulation was employed to investigate the interfacial thermal conductance between diamond and silicon substrate. The interfacial thermal conductance was computed based on Fourier's law. The simulation was done at different temperature ranges and results show that the interfacial thermal conductance between diamond-silicon is proportional to temperature and increases with temperature even above Debye temperature of silicon. Enhancement of thermal boundary conductance with temperature is attributed to inelastic phonon-phonon scattering at the interface. The system size dependence of interfacial thermal conductance was also examined. We found that thermal transport is a function of the system size when the size of system is smaller than the phonon mean free path and increases with the size of structure. We also simulated the effect of interface defect on phonon scattering and subsequently thermal conductance. The results also show that interface defect enhances acoustic phonon scattering which results in reduction of thermal boundary conductance. Our findings provide accurate and valuable information on phonon transport in diamond-silicon structure. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4775399>]

INTRODUCTION

Recently, due to the fast progress of fabrication technology and growing demands on the operation speed and economic feasibility of new electronic devices, there are a lot of interests in developing new materials and structures or composites with remarkable electrical and thermal properties to improve electrical performances of advanced electronic components.

Diamond-silicon structures attract a lot of attention as a composite substrate for application in electronic and optoelectronic industries to improve thermal issue.

In addition to the thermal management point of view, it has been reported that electron mobility of silicon embedded within acoustically hard material such as diamond can be enhanced via phonon engineering since the acoustic impedance of diamond is substantially higher than that of silicon.^{1,2} Diamond film on silicon substrate with properties similar to that of pure diamond crystal can be synthesized by chemical vapor deposition (CVD). In CVD method, diamond is deposited on silicon by applying a bias voltage between the silicon and microwave-frequency plasma.³⁻⁶ Heat conductance across diamond-silicon structure is one of the most important parameters that lead to optimize thermal management issue in electronic devices for increasing efficiency and life time of devices.

Molecular dynamic simulation has emerged as an enormous tool for the study of thermal transport in nanoscale materials. This method can complement both experimental and theoretical approaches by inclusion of anharmonic effects and atomic-level observations such as dislocation, defects, stressed boundaries that are not possible to control in experimental and theoretical works. There are two approaches to compute thermal transport by MD simulation. The first one is equilibrium MD simulation that is based on Green-Kubo formula,^{7,8} derived from linear response theory.⁹ Another approach is non-equilibrium molecular dynamics (NEMD) simulation which is based on Fourier's law.¹⁰ NEMD is first introduced by Chantrenne and Barrat¹¹ for measuring thermal conductivity in nanostructured materials. This method is most similar to the experimental measurements of heat conduction and is more suitable for computing thermal properties of inhomogeneous materials system and heat conductance across grain boundaries. Several simulations have done using NEMD on computing thermal conductivity of nanoscale systems and thermal conductance between dissimilar materials and across grain boundaries.¹²⁻¹⁷ In this method by making a temperature gradient across the structure, heat flux in the axial direction is related to the axial temperature gradient through thermal conductivity and thermal boundary conductance.

In the present work, we use NEMD to model thermal transport at the interface of diamond/silicon structure. This modeling aims to delineate effect temperature, system size, and of interface imperfection on phonon transport between diamond/silicon.

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SIMULATION DETAILS

In MD simulation, each atom is modeled as a point mass and interacts with other atoms through interatomic potential. The position and momentum of atoms are updated based on Newton's equation of motion. The setup of our simulation is shown in Fig. 1(a), where 6904 atoms were contained in a 27.2 Å by 27.2 Å by 103.9 Å simulation cell in the form of a diamond/silicon composite structure. Out of these atoms, 4704 were ordered in diamond structure and the rest formed as a silicon structure. Silicon side had $5 \times 5 \times 11$ unit cells and diamond side had $7 \times 7 \times 12$ unit cells. Brenner potential¹⁸ is used to model the interaction between Si-Si, C-C, and Si-C atoms. The interface of diamond/silicon was oriented along [001] crystallographic plane. Periodic boundary conditions were employed in x,y direction and fixed boundary condition applied at the extreme ends of structure in z direction (normal to the interface). In this simulation, the samples are initially set to a constant temperature to reach the equilibrium state with coupling time of 0.5 fs for 0.5 ns. The temperature of the system is controlled by velocity rescale algorithm. After the steady state condition is reached, Nosé-Hoover^{19,20} thermostats are set up at both ends of structure to induce temperature gradient in axial direction of structure for 10 ns. So, a constant amount of energy is added to the hot bath and the same amount of energy removed from the cold bath as described by Ikeshoji and Hafskjold.²¹ The baths encompass four unit cells along z-direction of the domain. The heat current through the system is determined as

$$J = \frac{Q}{A\Delta t}, \quad (1)$$

where Q is the thermal energy added to system and removed, A is surface area of interface, and Δt is the simulation time step.

Fig. 1(b) shows the steady state temperature profile along the structure. The stability in local temperatures was achieved after time period of 10 ns or 20 million time steps.

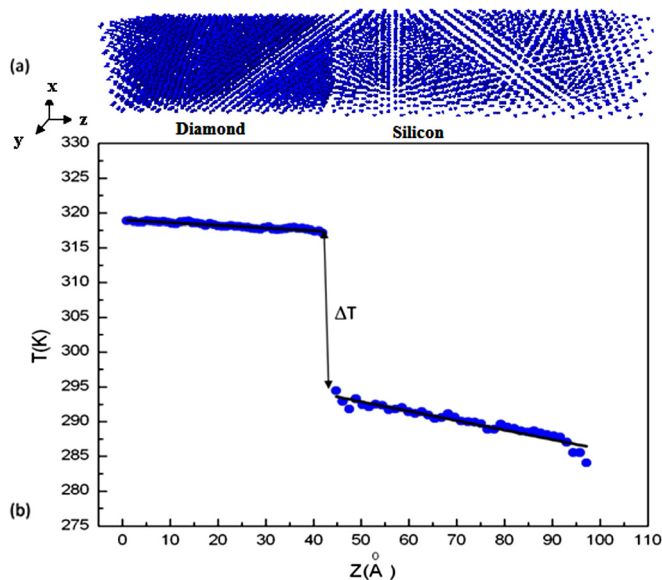


FIG. 1. (a) Schematic diagram of diamond-silicon nanostructure. (b) Temperature profiles across diamond-silicon structure.

Time-averaged smooth temperature profiles are extracted over the last 1 ns period. Local temperature in each layer of the system is given according to the atomic kinetic energy as

$$T_n = \frac{1}{3N_n k_b} \sum_{i=1}^{N_n} m_i V_i^2. \quad (2)$$

In the above equation, k_b is Boltzmann constant, N_n is the number of atoms in an atomic monolayer, and m_i and V_i are mass and velocity of i th atom in the layer, respectively.

Linear curve fitting using least square method applied to the temperature profile on each side. From the temperature profile, it is evident that discontinuity occurs at the interface of diamond-silicon, which is due to the mismatch of lattice-vibrational spectra and phonon velocities of two media.

Interfacial thermal conductance across the interface is defined as

$$G = \frac{J}{\Delta T}, \quad (3)$$

ΔT is temperature jump at the interface.

RESULT AND DISCUSSION

To provide a comparison to our MD simulation results, first we review a few experimental^{22–24} and theoretical²⁴ works performed on measuring thermal boundary resistance at the interface of diamond/silicon. Goodson *et al.*²² reported on thermal boundary resistance between silicon and diamond. They measured thermal resistance of diamond on silicon structure for the range of film thickness and reported that total resistance for the thinnest layer is upper limit of thermal boundary resistance between diamond/silicon, which is approximately 1.5×10^{-8} m²K/W. Klokov *et al.*²³ have shown that thermal boundary resistance between diamond/silicon with a thickness of 3 μm is 5.5×10^{-8} m²K/W. Goyal *et al.*²⁴ measured thermal boundary resistance between ultrananocrystalline diamond film with thickness 1.7 μm and silicon using experimental method: transient plane source and theoretical model: DMM and AMM. They showed that thermal boundary resistance obtained with experimental method at room temperature is 8.9×10^{-7} m²K/W that is three orders of magnitude larger than the value calculated with DMM and AMM by Goyal *et al.*²⁴

Our simulation result of thermal boundary resistance between diamond/silicon with perfect interface at room temperature is 2.98×10^{-9} m²K/W, which is an order of magnitude larger than theoretical predicted value by DMM and AMM. It is expected that MD result show higher thermal boundary resistance than DMM and AMM. In comparison with experimental values reported by Goodson²² and Klokov,²³ the computed value in this work is one and two orders of magnitude smaller. One of the possible reasons of discrepancy between experiment and MD simulation values could be due to interface imperfection. In MD simulation, interface is fully perfect but in reality the interface is not perfect. Synthesis and processing of real sample could result in various interface defects or intermixing that has significant effect on phonons transport. The phonons scattered by these

defect at the interface and thus thermal boundary resistance enhances. The effect of vacancy on thermal boundary conductance will be investigated in the interface defect effect section.

Temperature effect

Thermal boundary conductance between diamond/silicon in the temperature range of 150–900 K is calculated and shown in Fig. 2. In order to examine the effect of temperature on thermal boundary conductance, the system is first thermally equilibrated at each temperature point. As seen in the Fig. 2, by increasing the temperature, thermal boundary conductance increases and follows a similar trend above Debye temperature of silicon, around 645 K. The most likely explanation for this observation is that above the Debye temperature of the lower Debye temperature material at the interface, phonon scattering process becomes a²⁵ function of phonon population. Since phonon population above Debye temperature is driven classically, therefore, the number of phonons, which contribute in thermal transport, increases by increasing the temperature. This effect is attributed to inelastic phonon scattering that occurs when temperature increases and more phonons are excited, so there is a possibility for phonon with higher frequency at one side to scatter into phonons with lower frequency and transmit to the other side. It must be noted that only MD simulation in classical limit (above Debye temperature) is able to show inelastic phonon scattering. This effect has been observed by several MD simulations of thermal boundary conductance at the interface of solid-solid structure.^{12–14,16}

Size effect

It has been studied that thermal conductivity of diamond²⁶ and silicon²⁷ film is size dependent when the system size is smaller than mean free path of phonons. This effect is due to ballistic travel of phonons in the system with the size smaller than the phonon mean free path. Phonons which propagate ballistically have less contribution to thermal transport; therefore, thermal conductance would be a system size dependent.

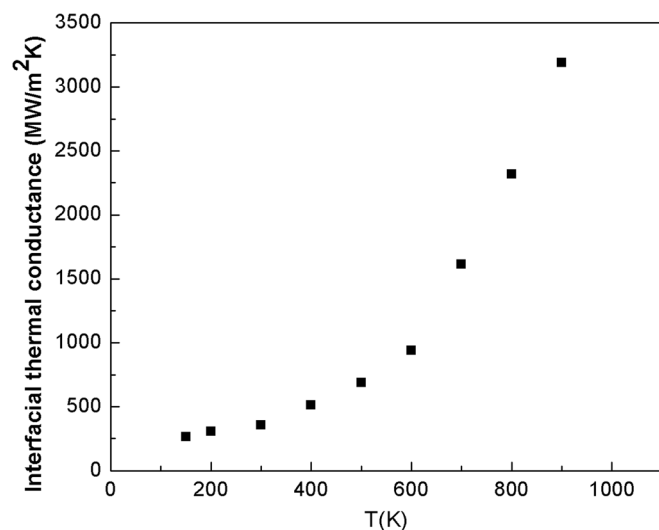


FIG. 2. Temperature dependence of thermal boundary conductance at diamond/silicon.

It was shown that thermal boundary conductance increases with the system size and then must saturate to the stable value once the system size reaches to the phonon mean free path in each slabs.²⁸ The length of the phonon mean free path in diamond and silicon is of the order of micrometers and simulating a system with the size in order of micrometer is limited due to computational resources.

However, in this work, we examined size dependence of interfacial thermal conductance between diamond/silicon for nanometer-sized systems which is smaller than the length of the phonon mean free path. Variation of thermal boundary conductance between diamond/silicon at 300 K versus size of the structure is depicted in Fig. 3. It is evident that by increasing the size of the system, thermal boundary conductance increases. This effect could be validated by accurate continuum model for heat conduction between two slabs. In this technique, interface acts as another material with a thermal resistance that depends on the entire system size.²⁸

Interface defect effect

Most of the computational and studies on thermal transport at solid-solid interface have considered perfect interface, but the interfaces in real materials are not atomically perfect.

In several experimental^{29,30} and theoretical works,^{12,31–33} effect of defect and interface on thermal conductance at the interface has been investigated. They found that interface defect has a strong influence on thermal transport at the interface of similar and dissimilar materials.

In this effort, effect of vacancy defect at the interface on the thermal conductance across diamond/silicon has been computed. A vacancy is random empty position in lattice structure and is made by taken M atoms away from interface atoms. The simulation method is the same as done for perfect interface. Fig. 4 shows that the thermal boundary conductance decreases with the increasing of vacancy density.

In order to provide more information on phonon mode scattered at the interface, vibrational density of state (VDOS) of phonon in diamond structure with and without

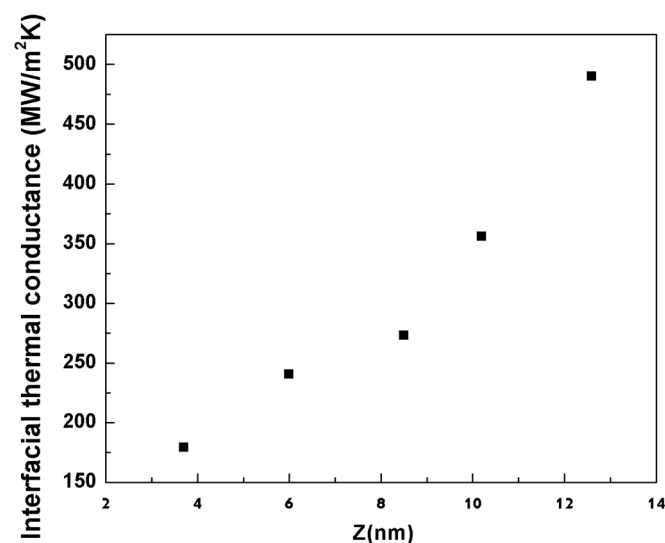


FIG. 3. Size dependence of the interfacial thermal conductance at diamond/silicon interface. The system temperature was 300 K.

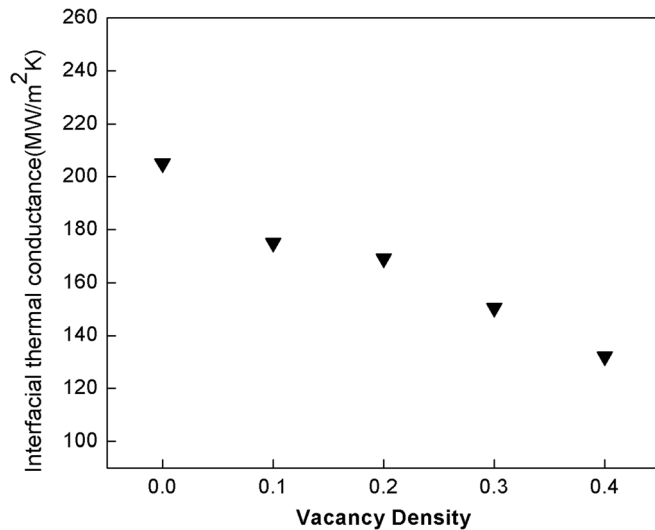


FIG. 4. Dependence of interfacial thermal conductance structure with length 37 \AA upon the density of the vacancy. The system temperature was 300 K.

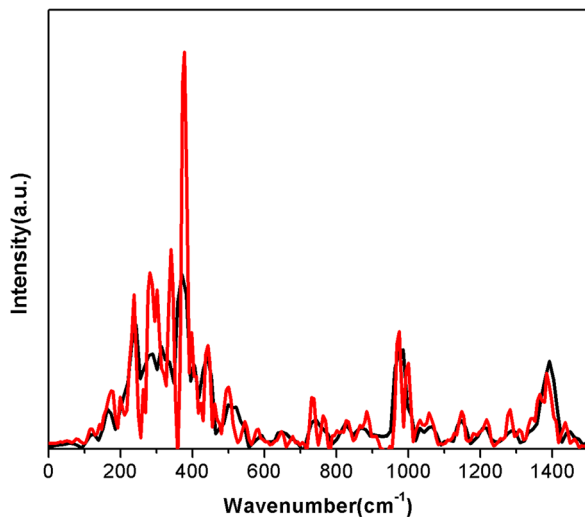


FIG. 5. Vibrational density of states in diamond at the interface of diamond/silicon structure.

vacancy at the interface has been calculated by conducting a Fourier-transform on the velocity autocorrelation function.³⁴ Strengthening or weakening of the different modes reveals more information about phonon scattering. Local DOS (LDOS) at the position of an interfacial diamond atom with and without vacancy is shown in Fig. 5. LDOS plot shows more changes in the intensity of VDOS within the frequency range of $300\text{--}600 \text{ cm}^{-1}$ which indicates more scattering in the long-wavelength acoustic phonons. Since acoustic phonon is dominant carrier of heat, it could be explained that the reduction in thermal boundary conductance in the presence of vacancy defect at interface is attributed to the increase of scattering in acoustic phonon at defect sites. These results are in agreement with the discrepancy between MD simulation result and experimental result.

CONCLUSION

In conclusion, we have examined the temperature, size of system, and interface defect on thermal boundary

conductance between diamond and silicon using molecular dynamic simulation. We explored the temperature range $T = 150\text{--}900 \text{ K}$, and our result indicates that thermal boundary conductance increases with increasing temperature, which suggests that inelastic phonon scattering happens above Debye temperature of silicon. The thermal boundary conductance is found to scale linearly with size whenever the system size smaller than the mean free path of phonons. The obtained result indicates that interface defect has strong influence on the thermal boundary conductance. In analysis of vibrational density of state of system with and without interface defect, we found that long wavelength acoustic phonons are scattered more strongly in the system with interface defect.

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