Enhancement of Interfacial Thermal Conductance of SiC by Overlapped Carbon Nanotubes and Intertube Atoms

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A new way was proposed to enhance the interfacial thermal conductance (ITC) of silicon carbide (SiC) composite through the overlapped carbon nanotubes (CNTs) and intertube atoms. By nonequilibrium molecular dynamics (NEMD) simulations, the dependence of ITC on both the number of intertube atoms and the temperature was studied. It is indicated that the ITC can be significantly enhanced by adding intertube atoms and finally becomes saturated with the increase of the number of intertube atoms. And the mechanism is discussed by analyzing the probability distributions of atomic forces and vibrational density of states (VDOS). This work may provide some guidance on enhancing the ITC of CNT-based composites. [DOI: 10.1115/1.4035998]

Keywords: interfacial thermal conductance, silicon carbide, overlapped CNTs, intertube atoms

1 Introduction

Silicon carbide (SiC) is emerging as a potential high-performance material which can be applied in electronics, nuclear reactors, aerospace, etc. [1–5], because of its wide band gap, large breakdown field, good mechanical property, and high thermal conductivity [1,6,7]. In the applications of high-temperature environment, it is especially required to estimate the heat dissipation in SiC devices and composites, where the thermal interface resistance dominates in phonon propagating. Recently, the studies on thermal transport through the interface have received more and more attention [8–18].

Carbon nanotubes (CNTs) have been theoretically predicted and experimentally proved to have ultrahigh thermal conductivity along the axial direction [19–25]. Therefore, CNTs are currently investigated as thermal interface materials both theoretically and experimentally [26,27]. It was simulated by molecular dynamics (MD) that only using pure CNTs to connect two solid surfaces as thermal interface material [28]. However, this interface structure is actually inconvenient for experimental implementation, because CNTs are grown on only one solid surface and cannot form a uniform channel between two solid slabs. In previous experimental study, CNT arrays were, respectively, grown on each SiC surface and then formed a composite sandwich structure, which has a low thermal conductivity as 0.5 W/m K [29]. Although a single CNT has a good thermal transport property, the thermal conductivity of CNT-based composites is still low due to the poor interfacial thermal conductance (ITC) among CNTs by van der Waals (VDW) forces.

Here, we proposed a new SiC–CNT array composite structure as SiC/CNTs/SiC. As shown in Fig. 1(*a*), two CNT arrays with different diameters are, respectively, grown on each SiC surface, so that the two SiCs are bridged by overlapped CNTs. Besides, intertube carbon atoms are added with covalent bonding between two CNTs. Some previous studies show that it is possible to construct the overlapped CNTs' structures in experiments. First, CNT arrays with different diameters could be easily grown on the surfaces of two SiC slabs [29]. And some molecular structures or polymer chains were able to be put inside CNTs in real experiments [30,31]. Moreover, the technique of joining of two single CNTs has been reported [32]. These related experimental studies imply that the structure of overlapped CNTs could be possibly realized in the future.

In this study, the ITC of SiC/CNTs/SiC composite was investigated by molecular dynamics simulations. We first gave a description of the model and simulation procedures. Second, the dependence of ITC on different numbers of intertube carbon atoms (N) was shown. Third, the temperature dependence of ITC was also studied. Finally, we performed the mechanism analysis through the probability distributions of atomic forces and vibrational density of states (VDOS).

2 Simulation Methods

Classical nonequilibrium molecular dynamics (NEMD) method is employed to study the ITC of SiC/CNTs/SiC using large-scale atomic/molecular massively parallel simulator (LAMMPS) package [33]. The interatomic bonding interactions within SiC and CNTs are described by Tersoff potential [34,35] including both two-body and three-body potential terms, which has been widely used to study the thermal properties of SiC and CNT [6,36,37]. And the interaction between SiC and CNT is the covalent bonding which is also described by Tersoff potential. In addition, the nonbonded interactions between overlapped CNTs are described by the Lennard-Jones potential

$$V_{\mathrm{LJ}}(r_{ij}) = 4\varepsilon[(\sigma/r_{ij})^{12} - (\sigma/r_{ij})^{6}] \tag{1}$$

where ε is the depth of the potential well, σ is the finite distance at which the interatomic potential is zero, and r_{ij} is the distance between atom i and j. The Lennard-Jones parameters are $\varepsilon_{\mathrm{SC-PC}} = 0.0028$ eV, $\sigma = 3.4$ Å, and the cutoff distance is set as 8.5 Å.

The simulation system consists of three parts (as shown in Fig. 1(*a*)), including the SiC of $4 \times 4 \times 4$ unit cells at both ends and the overlapped CNTs in the middle. The diameters of inner-tube and outer-tube of CNTs are 0.678 nm and 1.356 nm, respectively, corresponding to the chirality of (5, 5) and (10, 10). The atoms at boundaries in the longitudinal direction are fixed, and the

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periodic boundary conditions are applied in the other two directions. The simulation procedures begin by equilibrating the system at 300 K with a Nose–Hoover thermostat for 50 ps. Then, the hot region is raised to 320 K, and the cold region is lowered to 280 K using Langevin thermostat for 2.5 ns. Finally, it runs 5.0 ns to calculate the local average temperature and heat flow. The time step is set as 0.5 fs. The heat flux is deduced by tallying the energy added to the "hot reservoir" and removed from the "cold reservoir." The ITC is calculated by the ratio of heat flux to the temperature difference

$$G = J/\Delta T \tag{2}$$

The intertube carbon atoms are randomly distributed along both axial and circumferential directions of the overlapped CNTs. We use a combination of time and ensemble sampling to obtain better average statistics. The result of each N represents an average of five independent simulations with different random distributions of intertube atoms.

Figure 1 shows a typical setup of the system and corresponding temperature profiles. The longitudinal view of the structure is shown in Fig. 1(*a*), and the corresponding cross section views are shown in the insets of Figs. 1(*b*) and 1(*c*). Thermal interfaces of the overlapped segment of CNTs and the whole part between SiC are concerned in this work. The results of temperature profiles show that the temperature differences decrease obviously in the case of $N = 2$ compared with that in the case of $N = 0$, which indicates that the intertube atoms play a positive role in enhancing the interfacial thermal transport.

3 Results and Discussion

Figure 2 illustrates the effects of the number of intertube carbon atoms on the ITC between two CNTs (G_{CNTs}) and the total ITC of the system (G_{Total}). It is found that the ITC increases with the number of intertube atoms. The intertube atoms bridge the inner and outer CNTs through covalent bonds and form efficient channels for better heat transport, which lead to a sharp increase of ITC. Especially for N varying from 0 to 1, the ITC between two CNTs is enhanced by almost 10 times. It also indicates that the ITC finally becomes saturated with the increase of N. Since more atoms are concentrated at the overlapped segment of CNTs, the additional intricate channels may constitute a "traffic jam" which is not in favor of heat transport. On the one hand, the added intertube atoms provide more phonon transport channels, while on the other hand, intricate channels result in more phonon scattering.

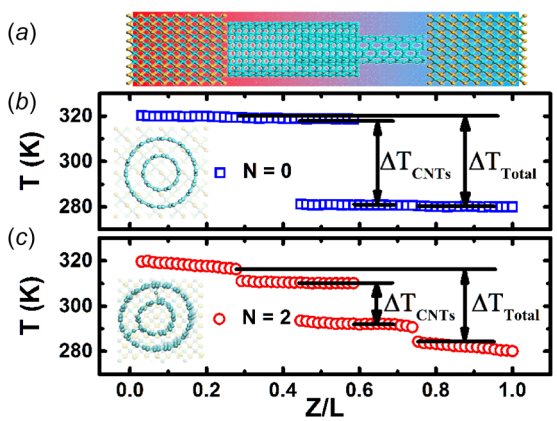

Fig. 1 (*a*) Longitudinal view of simulation system and (*b*) and (*c*) temperature profiles and cross section views of simulation system for the cases of $N = 0$ and $N = 2$. N denotes the number of intertube atoms. The overlapped segment of CNTs and the whole parts between SiC are considered as the thermal interfaces of two CNTs and the whole simulation system, respectively.

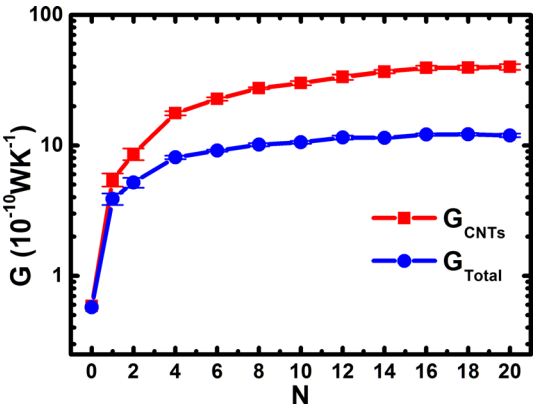

Fig. 2 The interfacial thermal conductance (G) between two CNTs (G_{CNTs}) and the total thermal conductance (G_{Total}) of simulation system at room temperature as functions of the number of intertube atoms (N). The interfacial thermal conductance shows a sharp increase from $N = 0$ to $N = 1$. Both G_{CNTs} and G_{Total} converge gradually with the increase of N. Finally, G_{CNTs} is enhanced by 2 orders of magnitude, and G_{Total} is enhanced by almost 20 times as well.

The two opposed effects cancel each other out, which leads to the saturation of ITC. Finally, compared with the case without intertube atom, G_{CNTs} can be increased by almost 2 orders of magnitude, and G_{Total} can be also improved by 20 times.

The enhancement of ITC by intertube atoms is a comprehensive result of two competing effects. The first effect is that a high-efficiency channel and more phonon modes along radial direction are introduced by intertube atoms, which contribute to the heat transport, thus increase intertube thermal conductance. The second effect is that the additional phonon modes and defects also open up new routes for phonon scattering, thus decrease the thermal conductivity of CNT and SiC [38]. In the case of SiC/CNTs/SiC, the main hindrance of thermal conductance comes from overlapped CNTs interacted by VDW forces. Compared with the effect of boundary scattering and mismatch of intertube phonon modes, phonon scatterings induced by additional phonon modes are negligible in SiC/CNTs/SiC. Therefore, the first effect of more phonon modes introduced by intertube atoms is dominating and conducive to phonon transport, which leads to the increase of ITC. Furthermore, the fact that thermal conductance increases dramatically and then converges as more intertube atoms are added, also confirms that the first effect of more phonon modes is stronger than the secondary effect of phonon–phonon scattering.

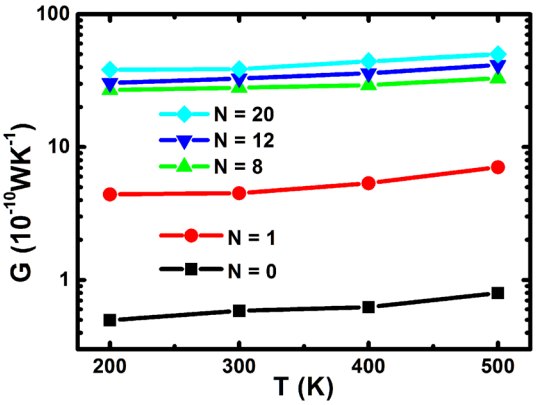

Fig. 3 The temperature dependence of interfacial thermal conductance (G) between two CNTs for some typical cases of different N. Monotonic increases are observed as a function of temperature for all the cases.

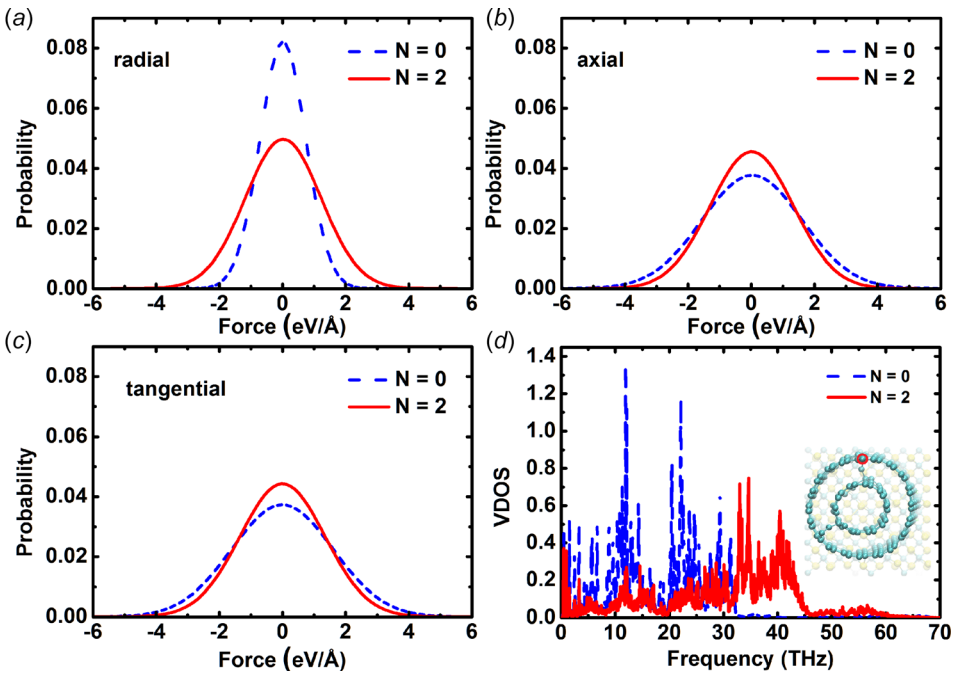

Fig. 4 The probability distributions of atomic forces along (*a*) radial, (*b*) axial, and (*c*) tangential directions, and (*d*) vibrational density of states (VDOS) along radial direction of the atom at the connection of outer CNT (circled one in (*d*)) for the cases of $N = 0$ and $N = 2$

The temperature dependences of ITC between two CNTs are shown in Fig. 3. A monotonic increase is observed in the ITC between two CNTs as a function of temperature. This is in striking contrast to the thermal conductivity behavior of CNTs and SiC, where the thermal conductivity at higher temperatures is found to be lower. The monotonic increase of ITC is also observed in previous work [28]. It might be due to the fact that as the temperature increases, more phonons are excited in both SiC and CNT, and thus, contribute more to the ITC. In general, at high temperatures, the phonon–phonon scattering begins to dominate, thus hinders phonon transport in SiC and CNT. In the current case, however, the intertube thermal transport is considerably poorer than that in CNT and SiC, higher temperature excites more phonon modes along radial direction, giving rise to a better phonon–phonon coupling at the interface.

In order to reveal the differences of atomic interactions for cases with and without intertube atoms, Fig. 4 shows the probability distribution functions of atomic forces along radial, axial, and tangential directions, and vibrational density of states (VDOS) along radial direction of the atom at the connection of outer CNT. Figure 4(*a*) presents an extension of atomic forces along radial direction for the case with intertube atoms. Covalent bonding induced by intertube atoms strengthens the interaction along radial direction, which is conducive to intertube thermal transport. By contrast, there are slight changes of axial and tangential atomic forces, as shown in Figs. 4(*b*) and 4(*c*). VDOS is an effective analysis method to reflect the change of phonon vibrations. Figure 4(*d*) suggests that more phonon modes along radial direction are excited due to the intertube atoms. Compared with the case without intertube atom, more phonon modes along radial direction are excited because of the strong covalent bonding induced by intertube atoms, which also contribute to enhance interfacial thermal conductance.

4 Conclusions

We proposed a new way to enhance the ITC of SiC composite through the overlapped CNTs and intertube atoms. By NEMD simulations, the most important result is that the ITC is enhanced dramatically by adding intertube atoms and converges with the increase of the number of intertube atoms. Compared with the case without intertube atoms, G_{CNTs} can be increased by almost 2 orders of magnitude, and G_{Total} can be also improved by 20 times. The probability distributions of atomic forces and the vibrational density of states indicate that the covalent bonding induced by intertube atoms strengthens intertube interactions and excites more phonon modes along radial direction, which leads to a better thermal transport. Our investigations may provide some guidance on enhancing the ITC of CNT-based composites.

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