



Thermal conductivity of defective graphene



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ABSTRACT

In this Letter, the thermal conductivity of defective graphene is investigated by using non-equilibrium molecular dynamics simulations. It is found that various defects including single vacancy, double vacancy and Stone–Wales defects can greatly reduce the thermal conductivity of graphene. The amount of reduction depends strongly on the density and type of defects at small density level. However, at higher defect density level, the thermal conductivity of defective graphene decreases slowly with increasing defect density and shows marginal dependence on the defect type. The thermal conductivity is found to become less sensitive to temperature with increasing defect density.

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1. Introduction

Graphene is a one-atom-thick planar sheet consisting of sp^2 -bonded carbon atoms in a honeycomb crystal lattice [1–4]. As the thinnest and strongest material ever discovered, graphene possesses remarkable properties that attract the attention of researchers worldwide. Different synthesis techniques such as graphitization of SiC surfaces [5] and chemical vapor deposition on Ni or Cu substrates [6] have been developed to produce graphene for potential applications. However, various defects such as vacancies and grain boundaries are unavoidable in the synthesized graphene [7]. The presence of defects can alter the mechanical and thermal properties significantly as reported in previous works [4,6–10]. However, most of the investigations hitherto focus on the single vacancy (SV) and its associated effects on the thermal conductivity (TC) [9,10] and on the stiffness of graphene under axial tensile loading [10,11]. The thermal properties of defective graphene with other defects such as double vacancies (DV) [7,12] and Stone–Wales (SW) [13,14] are still unclear. For example, which type of defects has a greater impact on the TC of graphene? How does the TC of defective graphene respond to the changes in the defect density and temperature?

In this work, we aim to explore the roles of SV, DV, and SW defects on the TC of graphene. The reverse non-equilibrium molecular dynamics (RNEMD) simulations are used to characterize the

thermal properties of graphene in the presence of different defects and defect densities.

2. Simulation model

The molecular dynamics (MD) simulations are performed using LAMMPS [15] in which the carbon–carbon bond interaction is described by the adaptive intermolecular reactive empirical bond order potential (AIREBO) [16]. This potential has been widely adopted for studying the properties of carbon-based nanomaterials [8,17–20]. The TC is obtained by using the RNEMD based on Muller-Plathe's approach [21]. The key idea of the RNEMD is to apply a heat flux to the graphene and determine the temperature gradient induced by this flux. Unlike RNEMD, non-equilibrium molecular dynamics apply a temperature gradient and then measure the resulting heat flux. Periodic boundary condition is applied in the plane (length and width directions). As shown in Fig. 1, the heat source and sink slabs are located at the middle and the two ends of graphene, respectively. A heat flux J (in unit of Watt) is imposed at these two sink slabs by exchanging the kinetic energies between the hottest atom in the heat sink slab and the coldest atom in the heat source slab in a microcanonical NVE ensemble (i.e., number of atoms, volume and energy are constant in the system). The exchange process is performed every 100 time steps. The total momentum and the energy of the system are conserved. The heat flux J due to the exchange of atoms is then given by

$$J = \frac{\sum N_{\text{transfer}} \frac{1}{2}(m_h v_h^2 - m_c v_c^2)}{t_{\text{transfer}}} \quad (1)$$

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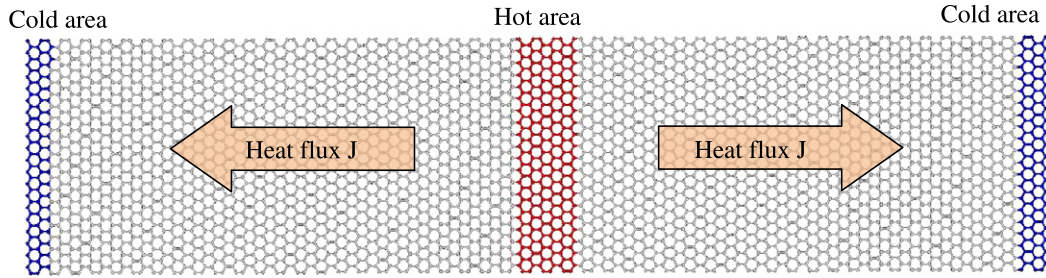


Fig. 1. Schematic of the simulation model for the non-equilibrium molecular dynamics.

where $t_{transfer}$ is the summation time, $N_{transfer}$ is the number of exchange, and subscripts h and c refer to the hottest and coldest atoms of which the velocities are interchanged.

Simultaneously, the temperature profile $T(x)$ with respect to the length is obtained after a time averaging over a prescribed time interval. The TC λ of graphene is then determined by using the Fourier law:

$$\lambda = \frac{J}{2A\partial T/\partial x} \quad (2)$$

where $\partial T/\partial x$ is the temperature gradient along the heat flux direction, and A is the cross-sectional area that the heat flux passes through. The factor 2 in the denominator is used to account for the periodicity of the system.

In the present simulation, the initial equilibrium configuration of graphene is achieved by the conjugate gradient minimization method. Then the system is equilibrated at the desired temperature in the NVT ensemble (constant number of atoms, volume and temperature) for 40,000 time steps with each time step of 0.5 fs. The environmental temperature is controlled by using the Langevin thermostat [22]. Thereafter, the heat flux is applied and the system is switched to NVE ensemble (constant number of atoms, volume and energy). Before the temperature profile is computed to infer TC, the system is allowed to evolve freely for 4×10^6 MD time steps during which the kinetic energies of the hottest atom in the hot slab and the coldest atom in the cold slab are exchanged at every 100 MD time steps. After reaching the steady state regime, the temperature gradient along the graphene length direction is obtained by averaging over 2×10^6 MD time steps. Unless otherwise stated, all the MD simulations are performed on the graphene with a length of 254.2 Å and a width of 60.9 Å with or without defects. The effects of different defects including SW, SV and DV on the TC are assessed by using the pristine graphene as the reference. All the defects are randomly distributed in the graphene. All the simulations are carried out at 300 K unless otherwise stated.

3. Results and analysis

In order to validate the computational model, we carried out simulations of defect-free graphene and carbon nanotubes (CNTs). The typical variation of the temperature obtained is shown in Fig. 2. The temperature profile is similar to those reported in previous works [8,17,18]. At the cold and hot ends, the temperature is nonlinear due to the finite size effects [8]. In order to avoid the edge effects, we use the linear middle portion (solid (red in the web version) line in Fig. 2) to extract the temperature gradient for the determination of TC by using Eq. (2).

We first carried out simulations on a 49.2 nm-long (10, 10) single-walled CNT to obtain its TC. By assuming the thickness of CNT as 1.42 Å [17,18], the TC is found to be 298.06 W/mK, which is in excellent agreement with 301.4 W/mK obtained by Xu and Buehler [17]. For the defect-free zigzag graphene nanoribbon (GNR) with a length of 20 nm and width of 2.1 nm, the TC ob-

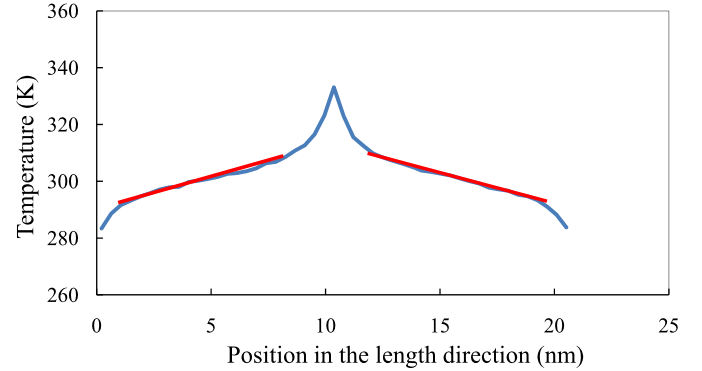


Fig. 2. Typical temperature profile in graphene along the heat flux direction.

tained by our computational model is 123.2 W/mK, which is close to the previously simulated value of 140 W/mK [18]. It is worth noting that the TC of CNTs and graphene measured in the experiments are in the range of 2500–5000 W/mK [23–25], which are much higher than the MD simulation results. This discrepancy may be attributed to the fact that the TC is strongly size-dependent as discovered by other researchers [26–30]. The CNTs or graphene investigated in experiments are micro meters long, whereas in the MD simulations only a few nanometers can be assessed due to the computational limitation. In order to confirm the size effect on TC, we carry out MD simulations at room temperature on a set of graphene with the same width but different lengths that vary from 8.4 to 200.8 nm. As shown in Fig. 3, the TC is approximately linearly dependent on the graphene's length L when L is smaller than 100 nm. Then the TC increases slowly with increasing L . By fitting the data points in Fig. 3, it is found that the TC follows a power law of $\lambda \sim L^{0.60}$ for graphene which is similar to the previous result of $\lambda \sim L^{0.47}$ obtained by Guo et al. [29] for armchair GNRs, although different models (graphene and GNRs) are simulated. It is reported by Haskins et al. [26] that TC of GNRs depends on the length (< 100 nm) and width (< 15 nm) linearly, then converge to a plateau. This length-dependent TC can be understood by noting that the mean free path of phonons in graphene is of the order of 775 nm [25], which is much larger than the size of the graphene in the MD simulations. Therefore, in addition to phonon–phonon scattering, there exists phonon scattering at the boundaries of the system, which consequently reduces the magnitude of TC obtained by MD simulations [9,10,12]. With regard to the width of graphene, phonons are free to travel across the simulation cell perpendicular to the heat flux (length) direction without scattering from boundaries due to the periodic boundary condition applied in the width direction [18,26,29]. Hence, the change in width of graphene will not influence the TC as significantly as the length. In view of the length effects, the TC obtained by MD simulation and experiment for graphene fails to agree with one another. The other possible reason for this disagreement is the

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