Grain size facilitating the heat transfer between graphene and silica substrate

Hossein Peer-Mohammadi, Ali Rajabpour*, Mansour Khanaki

Advanced Simulation and Computing Laboratory, Mechanical Engineering Department, Imam Khomeini International University, Qazvin 34148–96818, Iran

ABSTRACT

Grain size and thermal coupling to the substrate are among the major effects that may substantially dominate the heat transport properties of graphene-based nanodevices. In this study, we performed extensive transient molecular dynamics (MD) simulations on the basis of pump–probe technique to calculate the thermal boundary resistance (Kapitza resistance) between polycrystalline graphene and the silica substrate. As a remarkable finding, our results reveal that the thermal boundary resistance can be suppressed by decreasing of the grain size, so that the maximum thermal boundary resistance was predicted for the monocrystalline graphene. It was moreover observed that the thermal boundary resistance is less sensitive to the temperature variations for the polycrystalline graphene with smaller grain sizes. The obtained results and the corresponding underlying mechanisms were discussed by analyzing the roughness of polycrystalline graphene samples as well as the vibrational density of states. Considering the dominant role that the thermal boundary resistance plays in the nanoscale heat transfer, the findings of this paper may provide useful vision in heat management of graphene-based nanodevices.

1. Introduction

In graphene synthesize through chemical vapor deposition (CVD) method, polycrystalline graphene planes are usually produced instead of single-crystal graphene sheets. Structural defects at grain boundaries of polycrystalline graphene can significantly affect thermos-physical properties of graphene [1–5]. By decrease of grain size, the number of carbon atoms along the boundary will increase and consequently the density of structural defects will be enhanced. As a result, use of CVD method for polycrystalline synthesis graphene is expected to suppress the extraordinary thermal transport properties of graphene. Therefore, a fundamental understanding about the grain size effect on the thermal conductivity of graphene is of particular importance [6,7].

In fabrication of electronic devices such as photodetectors and transistors, two-dimensional structures have been used in contact with a substrate like silica. In these devices, excess heat generation increases the device temperature and consequently reduces its lifetime and efficiency. Therefore, such excess heat must be quickly and efficiently dissipated from the device. The rate of heat dissipation strongly depends on the thermal boundary resistance value between the two-dimensional plane and the substrate [8]. Therefore, understanding the heat transfer mechanism between the two structures has a significant effect on the optimal design of these devices.

Recently, it has been experimentally shown that the thermal properties of polycrystalline graphene is very sensitive to the existence of substrate [9]. In fact, grain boundaries play a critical role in heat transfer through graphene which was the subject of numerous researches in the past [10–14]. Bagri et al. investigated the effect of boundaries on graphene thermal conductivity using equilibrium molecular dynamics simulation and found that Kapitza resistance of the grains depends on the grain orientation [13]. Kotakoski and Meyer presented an atomistic model for generating polycrystalline graphene samples based on empirical observations [15]. Their model for polycrystalline graphene was later used for researches on mechanical and electrical properties of polycrystalline graphene as well [15,16]. Mortazavi et al. examined heat transfer in polycrystalline graphene samples in a multiscale approach. They used equilibrium molecular dynamics to examine the effects of grain size on the thermal conductivity of polycrystalline graphene. Their results showed that the thermal conductivity in ultra-fine polycrystalline graphene was lower than that of single-crystal graphene [17]. Hahn et al. calculated the thermal conductivity of graphene polycrystalline planes using the AEMD (approach-to-equilibrium molecular dynamics simulations) method [18]. Fan et al. showed that in the large graphene polycrystalline planes, the variation of the thermal conductivity with the grain size shows a dual behavior due to the different contribution of the in-plane and out-of-

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plane phonon modes [19].

Numerous studies have been conducted to calculate Kapitza resistance between two-dimensional nanostructures and substrates. Yue et al. recently reviewed both experimental works and molecular dynamics studies on Kapitza resistance of various types of two-dimensional nanostructures on substrates [20]. Zhang et al. [21] investigated thermal boundary resistance between two-dimensional silicene and silica substrate with both crystalline and amorphous forms using classical molecular dynamics simulations and a pump-probe method [22–25]. They reported the value of thermal boundary resistance of silicene/crystalline silica about 1.35×10^{-8} m^{2}K/W at 300 K. They also reported that the interfacial thermal resistance decreases by increasing the temperature of substrate. In another study, the value of 3.5×10^{-8} m^{2}K/W was reported for the thermal boundary resistance between graphene and silicon substrate at 300 K [26]. In 2017, Zhang et al. used MD simulation to investigate the thermal boundary resistance between phosphorous and crystalline silicon substrate and proved that the thermal boundary resistance of a silicon with a thickness above 3.12 nm is thickness-independent and only depends on the temperature [27]. Despite the huge difference between the thermal properties of crystalline graphene and amorphous graphene, there is no clear thermal boundary resistance between these two forms of graphene [28]. This is also the case for crystalline and amorphous silicon [29]. Farahani et al. [30] examined the thermal boundary resistance between MoS$_2$ and the silica substrate by transient molecular dynamics simulation. Their reported value for the thermal boundary resistance was interestingly consistent with the experimental value reported by Yalon et al. [31].

In this study, a transient MD simulation (inspired by the experimental pump-probe method) was carried out to calculate the thermal boundary resistance (Kapitza resistance) between polycrystalline graphene and the crystalline silica substrate. The size of grains in polycrystalline graphene are 2, 3, 5, 7 and 10 nm. Also, the effect of temperature with values of 300–500 K on the Kapitza resistance was investigated. The effects of the van der Waals interaction between graphene and silica substrate was also examined by changing the energy parameter of Lennard-Jones potential function. Roughness of polycrystalline graphene on the substrate and vibrational density of states were also calculated to explain the underlying physics of heat transfer.

2. Simulation details

In this research, LAMMPS package was utilized for all the simulations. Molecular dynamics is a powerful technique to calculate the thermal boundary resistance between nanostructures [32–35] since it can consider all harmonic and inharmonic effects as well as atomic structures of the interface. To determine the interactions between the carbon atoms in graphene and silicon and oxygen atoms in the silica structure, Tersoff potential function was used with parameters suggested by Monto et al. [36]. These potential coefficients have been widely used to calculate the interactions between silicon and oxygen in various Refs. [37–43]. For describing the van der Waals interaction between graphene and silica, Lennard-Jones (LJ) potential function was used as $U(r) = 4\chi\varepsilon[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^6]$. In this regard, χ specifies the strength of interaction between graphene and silica. Table 1 presents LJ parameters for different types of interactions [40]. The cutoff radius was also considered as 2.5σ in all simulations.

Table 1
LJ parameters between carbon atom in graphene and Si and O in silica substrate [40].

| Type of interaction	ε (eV)	σ (Å)
C–Si	8.909	3.326
C–O	3.442	3.001
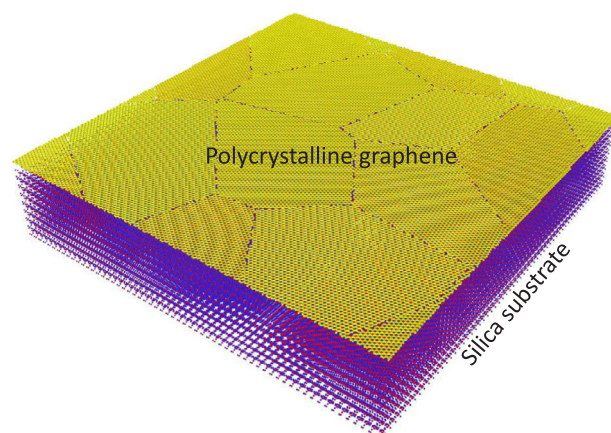
Fig. 1. Initial configuration of polycrystalline graphene with grain size of 10 nm on a crystalline silica substrate with thickness of about 6 nm. The length and width of the graphene are equals to 30 nm.

Graphene with a size of 30×30 nm^2 and crystalline silica with size of $30 \times 30 \times 6$ nm^3 were considered for initial atomic configuration of the system. Atomic positions of silica were generated utilizing VMD package. At the beginning of simulations, the graphene structure was placed 4 Å above the silica structure. Fig. 1 shows the initial configuration of the polycrystalline graphene with grain sizes of 10 nm on the silica substrate. In all simulations, periodic boundary conditions were assumed in two planar directions and free boundary condition was considered in the normal direction. The time step was set as 0.5 fs in all simulations.

3. Results and discussion

3.1. single-crystal graphene and validation

In this study, the pump-probe method was used to calculate the thermal boundary resistance between graphene and silica [23–25,30,44,45]. Transient MD technique has been widely employed to calculate the thermal boundary resistance [27,46–48]. At the start of simulation, providing a Nose-Hoover thermostat, the temperature of the system gradually increased from 0 to 300 K in 100 ps and then maintained for 25 ps in zero-stress conditions in an NPT ensemble with $T = 300$ K. Ultimately, NVE ensemble was used to reach the equilibrium state over a time period of 50 ps. At these conditions, the equilibrium distance between graphene and silica substrate was obtained about 3.2 Å. In order to start the pump-probe method, a heat pulse of $\dot{q} = 50 \times 10^{-4}$ W was applied to the graphene sheet in 50 fs as shown in Fig. 2. This heat pulse led to increase of the graphene temperature to 460 K. Then, by switching off the heat pulse, the graphene sheet started to exchange energy with the silica substrate in an NVE ensemble in 200 ps. During this time, the graphene temperature (T_{Gr}) and the silica temperature (T_{Si}), as well as the total energy of the graphene (E_t) were calculated and recorded. In order to reduce the statistical fluctuations, the temperature and energy were averaged every 100 steps. Fig. 3 shows the temperature variations of graphene and silica substrate
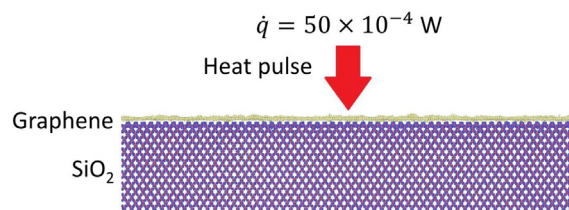
Fig. 2. The heat pulse value given to the graphene sheet during 50 fs in a pump-probe MD technique.