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Heat flow diversion in supported graphene nanomesh

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ABSTRACT

Redirection of energy carrier propagation by geometric confinement is studied through the analysis of in-plane and cross-plane thermal transport within various graphene nanomesh (GNM) configurations using molecular dynamics (MD) simulations. As the transport channel width decreases with an increase in porosity, the effect of redirection increases; thus, the in-plane thermal conductivity of large-porosity GNM is more dependent on hole arrangement. Since higher porosities weaken the GNM structure due to a larger population of broken bonds, carbon atoms within the graphene structures are more easily influenced by interactions with the substrate silicon (Si) block. Subsequently, increase in porosity leads to the decrease of interfacial thermal resistance. At higher porosities, lower interfacial resistance and in-plane thermal conductivity cause diversions (and redirections) in heat flow from the GNM to the underlying Si substrate. Our study suggests that this method of heat flow redirection can be applied as an effective means to control and manage heat transfer within numerous applications; extension to the improved conductivity calculation accuracy can also be achieved through the inclusion of this diversion analysis.

1. Introduction

Since graphene's first successful isolation [1], and the identification of its high electrical [2] and thermal [3,4] conductivity properties, these characteristics have made this novel material very promising for advancement and innovation within electronic applications [5–8]. Many research efforts have explored extensions of graphenes highly desirable attributes to more diverse and broader applications by manipulating its properties (e.g., boosting desirable and suppressing undesirable properties). Controlling thermal transport (i.e., direction and magnitude) is of great interest for energy conversion and electronic system design, with thermoelectric (TE) energy conversion being one such example of manipulation [9–12]. For optimal performance within TE devices, the reduction of thermal conductivity (k), while maintaining or enhancing the TE power factor ($\sigma_e S^2$, σ_e: electrical conductivity and S: Seebeck coefficient), is critical. Moreover, the direction of heat transfer can be controlled by altering thermal transport properties; utilization of such methods can be employed to enhance heat dissipation [13].

Previous efforts to manipulate the thermal conductivity of graphene have emphasized the use of chemical approaches to introduce defects and/or functional groups as an avenue to control resultant thermal properties [14–18]. Defects increase phonon scattering to thereby suppress thermal transport; however, defects are difficult to control and their effect on thermal transport is reported to be minor when defect concentration is large [19,20]. Moreover, defects parasitically degrade other desirable properties (e.g., electrical conductivity in TE applications) [14]. Edge passivation [21], isotope engineering [22], strain [23], and grain boundary [17,24] have also been considered for thermal transport control; among the various methods, the technique of geometrically modifying or constricting a graphene sheet is suggested as one of the most feasible and affordable methods [25] to effectively control thermal transport [26]. The use of graphene nanomesh (GNM), which contains nano-sized periodic holes on a graphene sheet, is one such example representative of the application of geometric constraints towards effective thermal transport control [27,28]. Due to their tunable bandgap through adjustment of pore size and periodicity, GNM presents itself as an excellent candidate for thermoelectric energy conversion [29] and field-effect transistors [6,7,27,30], energy storage [31], as well as gas detection [32,33].

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Recent studies utilizing molecular dynamics (MD) have primarily investigated free-standing GNM, whereas experimental studies and applications mainly employ GNM within supported/sandwiched form [27,32–34]. As the presence of a substrate is known to greatly influence thermal behaviors of single-layer graphene (SLG) and graphene nanoribbon in a dramatic way [12,35–39] studying GNM in a supported form provides more accurate insight to thermal transport behaviors within applications. In the study of supported GNM, Si is selected as a substrate material to achieve maximized heat diversion effect (due to its high thermal conductivity), and because of our continuing interest in Si-supported graphene [40] and wide engineering applications (e.g., detectors in aqueous environments [41], Schottky junction solar cells [42], and photonics [43]). Hole arrangement and porosity are controlled in suspended and Si-supported GNM; their effects on thermal transport, and the underlying physics of said effects, are examined.

2. Simulation methods

Periodic boundary conditions are applied in the *y*-axis direction, while fixed and free boundary conditions are used for the *x*- and *z*-axis directions (to avoid interactions between periodic cells), respectively. Here, *x* and *y* are in-plane directions, and heat flows in the *x* direction (Fig. 1a–c). A sample length (*l* in *x* direction) of 47.2 nm and a width (*w* in *y* direction) of 23.4 nm are selected to minimize lattice mismatch in Si/SLG heterostructures. The Si block length and width are of the same dimensions as the graphene sheet; the height of the Si block is 3.80 nm in the *z*-axis direction. Adaptive intermolecular reactive empirical bond order (AIREBO) [44] potential is used to model C–C bonded interactions, and the three-body Stillinger-Weber [45] potential is employed for modeling Si–Si interactions. Short-range van der Waals (vdW) interactions [46,47] between C and Si atoms are modeled by the 12–6 Lennard-Jones (LJ) potential, $\varphi_{LJ} = 4\varepsilon[(\sigma/r)^{12} - (\sigma/r)^{6}]$, where ε is the minimum energy, σ is the distance at which the intermolecular potential between the two particles is zero, and r is the distance of separation between particles. The potential parameters used in our simulations are based on the vdW interactions in the universal force field (UFF) model by Rappe et al. [48]. All simulations were performed using Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [49]; the total simulation time and time step size for all simulations are chosen to be 2.5 ns and 0.5 fs, respectively. In this work, reported results are the average of five independent simulations.

Two different arrangements of holes [square (SA) and zigzag (ZA)] are employed within this research (Fig. 1a and b). In each simulation, hole sizes are uniform and the radius ranges from 0 to 28.5 Å, while the hole center positions are fixed; thus, the porosity (of the simulation cell central region) is controlled up to 75%. With the maximum possible porosity being 78.5%, to avoid potential tearing and distortion of the graphene sheet structure, we select a porosity of 75% as the maximum investigated porosity size. With various hole arrangements and porosities, we investigate in-plane thermal transport in both stand-alone and Si-supported GNMs, as well as cross-plane transport in Si/GNM systems.

Thermal transport, characterized by conductivity and

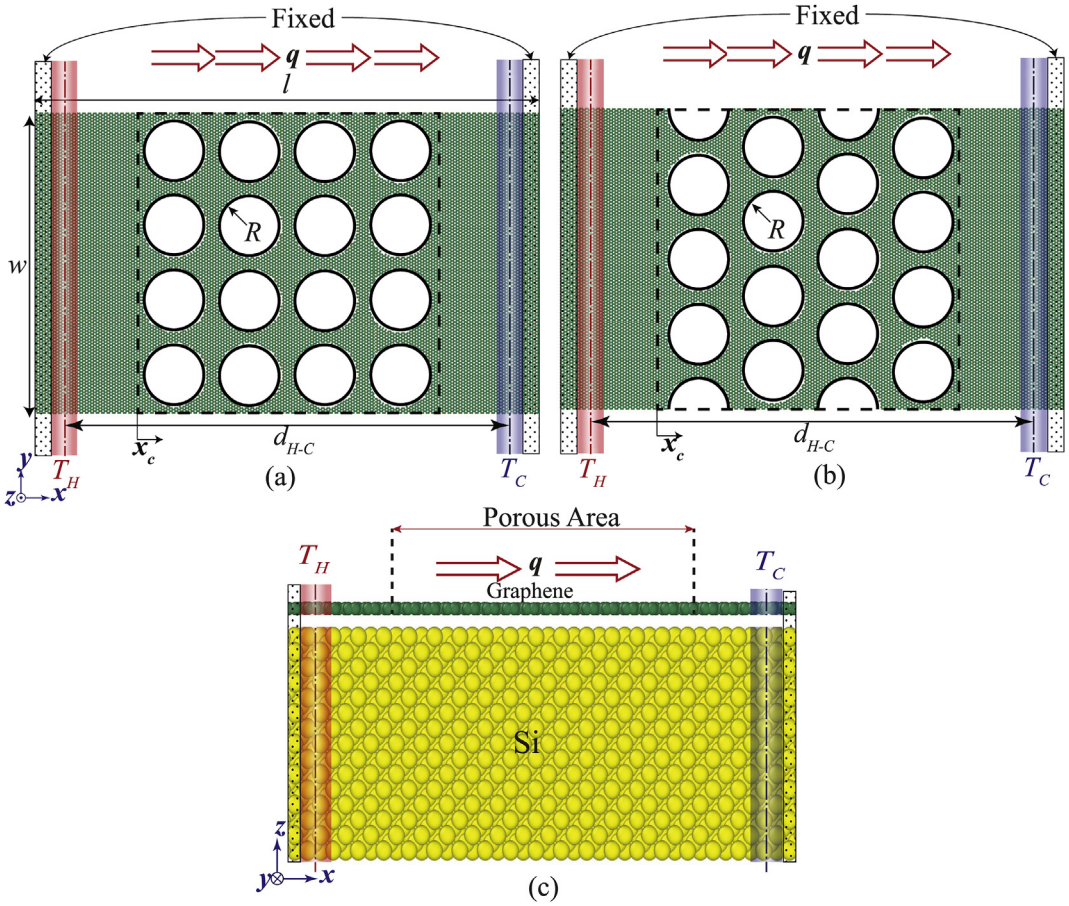

Fig. 1. Simulation models for the calculation of thermal transport properties. (a) Square hole arrangement and (b) zigzag hole arrangement of GNM. (c) GNM with Si substrate (side view). Separate hot and cold thermostats are placed on Si and GNM. (A colour version of this figure can be viewed online.)