Physics Letters A 380 (2016) 2105–2110

Contents lists available at ScienceDirect

Physics Letters A

www.elsevier.com/locate/pla

Triggering wave-domain heat conduction in graphene

Wen-Jun Yao, Bing-Yang Cao *

Key Laboratory for Thermal Science and Power Engineering of Ministry of Education, Department of Engineering Mechanics, Tsinghua University, Beijing 100084, PR China

ARTICLE INFO

Article history:
Received 16 March 2016
Received in revised form 12 April 2016
Accepted 16 April 2016
Available online 19 April 2016
Communicated by R. Wu

Keywords:
Non-Fourier heat conduction
Graphene
Mechanical wave
Molecular dynamics simulation

ABSTRACT

Using non-equilibrium molecular dynamics simulations, we systematically investigate the non-Fourier heat conduction in graphene under steady high heat flux. The results show that if two triggering factors, i.e. steady high heat flux and tensile stress, are satisfied simultaneously, a low-frequency mechanical wave and corresponding wave-like energy profile can be observed, which are distinctly different from ripples and linear temperature profile of the normal Fourier heat conduction. This mechanical wave provides an additional channel of heat transport and renders graphene more conductive without changing its pristine thermal conductivity. What's more, as the heat flux or original bond length increases, its frequency increases and energy transported by this mechanical wave is also on the rise. Further analyses show that such anomalous phenomenon is not arising from the high-energy or high-frequency pulses and also not artifacts of the velocity-exchange method. It is a dissipative structure, a new order state far from thermodynamic equilibrium, and the corresponding nonlinear relationship between the gradient of the wave-like kinetic temperature and the heat flux enables more efficient heat transport in graphene.

1. Introduction

The combination of high-efficiency heat dissipation and low-handling thermal management makes graphene a promising candidate material for energy, electronics, and optical applications [1–4]. In the past decade, large amount of studies have been dedicated to heat transport in graphene [1,3–20], considerable part of which deals with its reducible thermal conductivity under low heat fluxes, where phonons are blocked or scattered by defects [11–14], dopants [11,12,15,16], strain [17,18] and so on [19,20]. These studies show a tunable thermal conductivity of over 25 times and a high thermoelectric figure of merit ZT of 2∼3 [12,14,19], making graphene possible for thermoelectrics. But for high power or high frequency applications, the heat dissipation, manipulation or even enhancement of thermal conductivity of graphene at high heat fluxes is still a challenge.

Earlier studies demonstrated that in low dimensional materials, the non-Fourier heat conduction is easy to be activated by instantaneous high heat fluxes [21–23]. For example, if imposing a local heating with a duration of 1 ps into graphene, a thermal wave, symbol of non-Fourier heat conduction, would be observed [23]. Likewise, non-Fourier heat conduction could also occur at steady high heat fluxes. Recently, Zhang et al. [24] first reported that as the steady high heat flux exceeds a critical value, a low-frequency transverse acoustic wave will be excited in single-walled carbon nanotubes (CNT), which results in an anomalous temperature profile and transports energy effectively. Similar mechanical waves were also observed in graphene nanoribbons and hybrid GE/SE monolayers under steady high heat flux conditions [25,26]. This non-Fourier heat conduction mechanism suggests a totally fresh idea of solving heat dissipation. But this is only the beginning. How to trigger this wave-domain transport mode still deserves to be investigated.

Here, we demonstrate the wave-domain heat transfer mechanism under steady high heat fluxes again in graphene. And by systematically analyzing the influences of the bond length, heat flux, boundary conditions, system length and simulation parameters on the frequency of the mechanical wave using a non-equilibrium molecular dynamics (NEMD) method, we find two triggering factors of this anomalous heat conduction phenomenon, that is, the heat flux and the strain, and further we discuss the underlying mechanism of the wave-domain heat conduction.

2. Simulation detail

In this work, we systematically investigate the non-Fourier heat conduction of the zigzag-edged graphene sheet under steady state in models with Brenner potential [27], by using NEMD method [10,14,23,28,29]. In the MD simulations, periodic boundary conditions are used along the length direction, and as for the width direction,

* Corresponding author. Fax: +86 10 6279 4531.
E-mail address: caoby@tsinghua.edu.cn (B.-Y. Cao).

http://dx.doi.org/10.1016/j.physleta.2016.04.024
0375-9601/

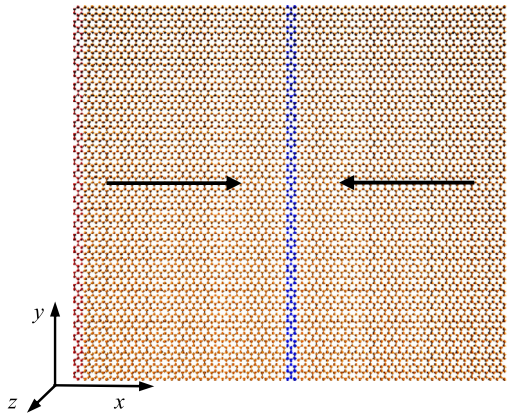

Fig. 1. Schematic of simulation system including a high-temperature slab (red) and a low-temperature slab (blue). (Color online.)

we performed free and periodic boundary conditions respectively and no qualitative changes are found on the conclusions.

As illustrated in Fig. 1, the heat current is from the high-temperature slab (the red atoms on the side) to the low-temperature slab (the blue atoms in the middle), which is imposed by using the velocity exchange method developed by Müller [30] and calculated from Eq. (1):

$$J = \frac{\sum_{transfers} \frac{m}{2}(v_h^2 - v_c^2)}{t}, \tag{1}$$

in which m is the atomic mass of carbon, v_h is the velocity of the hottest atom in the low-temperature slab, v_c is the velocity of the coldest atom in the high-temperature slab, t is the statistical time. Total energy and momentum of the system are conserved during the velocity-exchange, while the system temperature is kept at 300 K with the Nosé–Hoover thermostat [31]. The atomic motion is integrated by the leap-frog scheme with a fixed time step of 0.5 fs. Each simulation case runs for 1 ns to reach a steady state, and then for 1.5 ns to average the temperature, heat flux, and bond-length over time. During the simulation, the system is equally divided into 50 slabs along the length direction, and the local instantaneous temperature for each slab, according to the energy equipartition theorem, is defined through the averaged kinetic energy as:

$$T_i = \frac{2}{3N_i k_B} \sum_{j=1}^{N_i} \frac{P_j^2}{2m}, \tag{2}$$

where N_i is the atom number of i-th slab, k_B is the Boltzmann constant and P_j is the momentum of the j-th atom.

3. Results and discussion

Imposing a small heat flux into graphene, heat is transported following Fourier's heat conduction law and a typical configuration is observed, as shown in Fig. 2. We know that the stability of two-dimensional (2D) materials has not been very clear yet [32–34]. From our simulations, the first thing to note is that the spatial distribution of carbon atoms is not perfectly two-dimensional. Ripples spontaneously appear under low heat fluxes. They look like randomly distributed and don't disappear with time, which is consistent with theoretical researches [35]. Thus, it is demonstrated that the graphene is not a strictly 2D crystal, and its existence doesn't contradict the argument that strictly 2D crystals are thermodynamically unstable and could not exist in a free state. Although 2D graphene embedded in a three-dimensional space has a tendency to be crumpled, the ripples are still suppressed by anharmonic coupling between bending and stretching modes, making graphene a quasi-2D structure.

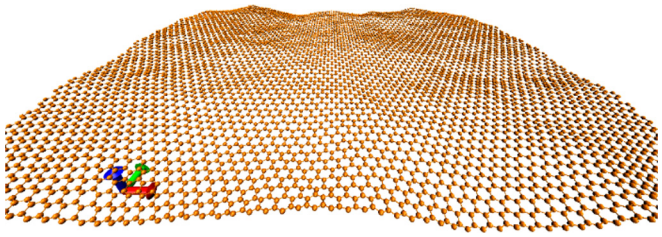

Fig. 2. Snapshot of ripples in simulation system.

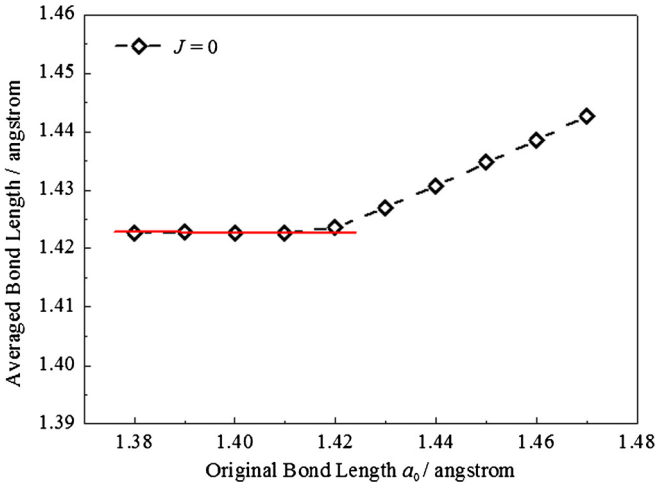

Fig. 3. Averaged bond length $\bar{a}$ of systems with a heat flux $J = 0$ versus original bond length a_0. (Color online.)

Additionally, ripples also introduce strain into the simulation systems. Fig. 3 shows the averaged bond length $\bar{a}$ of systems with a heat flux $J = 0$, calculated after reaching a steady state, as a function of the original bond length a_0, set before simulation. It can be seen from the red line in Fig. 3, as the original bond length increases from 1.38 Å to 1.41 Å, the averaged bond length stays the same and is always larger than the original one, suggesting that systems with bond length equal to or smaller than 1.41 Å are initially compressed. But because no restrictions are on the out-of-plain direction in graphene due to its quasi-2D structure, the graphene sheet can only be crumpled while its bond length cannot be decreased. However, with larger original bond length, the averaged bond length increases gradually, indicating that the systems with the original bond length of 1.42∼1.47 Å are stretched to different degree at the steady state. Some researches give the unstrained bond length of graphene is 1.42 Å [36–38], which is in accordance with our conclusion that no stress is in the systems with the bond length smaller than 1.42 Å, but because of the existence of the ripples mentioned before, the systems with the original bond length equal to 1.42 Å are also stretched in our simulations.

In our simulations, under a small heat flux, ripples spontaneously appear and heat is transported by phonons, i.e., the lattice vibrations, following Fourier's heat conduction law, in both strained and unstrained graphene. But when the heat flux exceeds a critical value, a mechanical wave may be observed, which is quite different from ripples. It provides an additional channel for the non-Fourier heat conduction and enables graphene more conductive, which we will mainly focus on in this paper. For example, we impose a heat flux of 2713 GW/m^2, high enough to excite the mechanical wave, into the pristine graphene. Then we calculate the temperature profiles and grape some images of the real system at different time, presented in Figs. 4(a) and 4(b).