Monte Carlo Simulation for Phonon Heat Transfer in A Graphene Ribbon

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Introduction

Graphene, a solitary layer of carbon iotas in a two-dimensional (2D) honeycomb cross section, shows remarkable electrical, thermal, and mechanical properties. Test estimations of room-temperature thermal conductivity from 2000 to 5000 W/mk [1]. Graphene has extremely high thermal conductivity contrasted with metallic heat conductors or other carbon-based materials, like graphite or diamond. The graphene nanoribbon is a type of graphene usually imagined in gadgets since it tends to be designed utilizing standard lithography [2]. Graphene nanoribbon can be created with different widths and lengths, just as various line-edge harshness [3]. Phonon transport in graphene nanoribbon can be adjusted by altering the aspects, inferable from line-edge-unpleasantness dispersing and ballistic transport features [4]. In addition, thermal flow in graphene nanoribbon is additionally tunable through the decision of substrate, isotope concentration, and ribbon crystal orientation [5]. Thermal conductivity designing in these frameworks has been drawing in impressive interest as of late.

Abstract

Phonons will be launched from either side of a graphene ribbon that is 1 micron long.

The phonons will interact with each other and physical boundaries. For this reason,

scattering of photon will take place. The graphene ribbon is suspended, does not interact with any other materials, which has no imperfections. For nanoscale, Fourier's Law is not applicable for heat transfer because of length scale [6]. Linearize Boltzmann relation can be used to solve the thermal conductivity problem, but we don't have adequate way to solve the scattering time value. 2D heat transfer model will be considered for this problem, as graphene is a 2-D material having its depth only h=0.335 nm. For heat transfer, only one longitudinal acoustic (LA) and one transverse acoustic (TA) phonon will be considered. By the set-up of equations and parameters, we use python to plot the graph which shows the conductivity varies as a function of ribbon width and temperature. We use Monte Carlo simulation which used to model the probability of different outcomes in a process that cannot easily be predicted due to the intervention of random variables to tract down the position and velocity with time. In this paper, the width of graphene nanoribbon is given which varies from 10 to 500 nm and the length is 1

micron meter in the problem statement. After the determination of the equation for thermal conductivity, the distribution of temperature and width would be calculated and shown by the python plotting.

Method and Simulation

Phonon Heat Conduction in Graphene

When the heat flux pass through a graphene atomic plane, the total heat flux can be multiplied by the system volume which is expressed by the following equation

$$\overline{W} = -\sum_{s,\bar{q}} \bar{v}(s,\bar{q})\hbar\omega_s(\bar{q})N\big(\bar{q},\omega_s(\bar{q})\big) = \sum_{s,\bar{q}} \bar{v}(s,\bar{q})\hbar\omega_s(\bar{q})n(\bar{q},\omega_s) \quad (1)$$

Where $\bar{v}(s,\bar{q})\hbar\omega_s(\bar{q})$ is the energy carried by one phonon, $\bar{v}(s,\bar{q}) = d\omega_s/dq$ is the phonon group velocity and $N(\omega,\bar{q}) = N_0(\omega,\bar{q}) + n(\omega,\bar{q})$ is the number of phonons in the flux.

The Umklapp scattering process take places after phonon moving around the graphene nanoribbon and when a phonon with the wave vector $\bar{q}(\omega)$ absorbs another phonon from the heat flux with the wave vector $\bar{q}'(\omega')$. The momentum and energy conservation laws for this type of scattering process can be written as

$$\bar{q} + \bar{q}' = \bar{b}_i + \bar{q}'' \quad (2)$$

$$\omega + \omega' = \omega'' \quad (3)$$

Where $\bar{b}_i$, i=1, 2, 3, is one of the vectors of the reciprocal lattice. The second type of process occurs when the heat flux phonons $\bar{q}$ decay into two phonons with wave vectors $\bar{q}'$ and $\bar{q}''$ leaving the state $\bar{q}$, or when two phonons $\bar{q}'(\omega')$ and $\bar{q}''(\omega'')$ merge together, forming a phonon with the wave vector $\bar{q}(\omega)$, which corresponds to the phonon arriving at the state $\bar{q}(\omega)$. This type's conservation laws are provided by

$$\bar{q} + \bar{b}_i{}' = \bar{q}' + \bar{q}'', \quad i = 4,5,6 \quad (4)$$

$$\omega + \omega' = \omega'' \quad (5)$$

Because the length scale in the graphene nanoribbon is small, the Fourier's law is no longer applicable under the conditions **[7]**, which leads us to the Boltzmann Transport Equation (BTE)

$$\frac{\partial f}{\partial t} + v\nabla_r f + \frac{F}{\hbar}\nabla_k f = \left(\frac{\partial f}{\partial t}\right)_c \quad (6)$$

The BTE provides the non-equilibrium distribution function of energy carriers, which requires that particles act independently of on another. The relaxation time approximation can be expressed by following equation

$$(\frac{\partial f}{\partial t})_c = \frac{f-f_o(T,E,\mu)}{\tau(r,x)} \quad (7)$$

Where $\tau(r,x)$ is the relaxation time, and $f_o$represents the equilibrium distribution of carriers. For the Umklapp process the combined scattering rate can be calculated as

$$\frac{1}{\tau_U(s,\bar{q})} = \frac{1}{\tau_U^I(s,\bar{q})} + \frac{1}{\tau_U^{II}(s,\bar{q})} \quad (8)$$

It is important to note that for small phonon wave vectors (long wavelength), $q \to 0$ equals the Umklapp-limited phonon lifetime $\tau_U \to \infty$. As a result, without an arbitrary truncation process, the intrinsic thermal conductivity cannot be calculated using solely Umklapp scattering. The boundary scattering term in graphene corresponds to scattering from the rough edges of graphene flakes. Because graphene flake is only one atomic layer thick and the phonon flux is parallel to the graphene plane, there is no scattering from the top and bottom sides.

Phonon Thermal Conductivity

In this section, there is a simpler model of the thermal conductivity of graphene, which originates from Klemens' approach to the thermal conductivity analysis of bulk graphite and graphene **[8]**. By introducing two Gruneisen parameters γ_s obtained independently for each of the heat conducting phonon polarization branches s, and for each phonon branch, keeping the velocities and cut-off frequencies separate. These modifications allowed us to represent the characteristics of graphene's phonon dispersion. Only one longitudinal acoustic (LA) and one transverse acoustic (TA) phonon are considered for heat transmission **[9]**. The thermal conductivity equation is as follows:

$$\kappa = \frac{1}{4\pi t k_B T^2} \sum_s \int_{k=0}^{k=max} v_s \, (\hbar\omega)^2 \tau_p \frac{e^{\hbar\omega_p/k_B T}}{(e^{\hbar\omega_p/k_B T}-1)^2} k dk \quad (9)$$

The cut-off frequency for Umklapp process cannot be introduced by analogy with bulk graphite. After introducing separate lifetimes for LA and TA phonons, the relaxation time calculation can be expressed by

$$\tau_{U,s}^K = \frac{1}{\gamma_s^2} \frac{M v_s^2}{k_B T} \frac{\omega_{s,max}}{\omega^2} \quad (10)$$

Where s=TA or LA,v_s is the average phonon velocity, T is the absolute temperature, k_B is the Boltzmann constant, $\omega_{s,max}$ is the maximum cut-off frequency and M is the mass of the unit cell. After combining the equation, the scalar thermal conductivity equation is expressed by

$$\kappa_S = \frac{M}{4\pi T h} \sum_{S=TA/LA} \frac{\omega_{smax} {V_s}^2}{{\gamma_s}^2} F \quad (11)$$

With F expressed by

$$F = \int_{\varsigma_{s\,min}}^{\varsigma_{s\,max}} \frac{\varsigma_s e^{\varsigma_s}}{(e^{\varsigma_s - 1})^2} d\varsigma \tag{12}$$

For calculation of $\varsigma_{s\,max}$ and $\varsigma_{s\,min}$, we use the following equations,

$$\varsigma_{max} = \frac{\hbar \omega_{s,max}}{k_B T}$$

$$\varsigma_{min} = \frac{\hbar \omega_{s,min}}{k_B T}$$

The minimum frequency can be calculated by

$$\omega_{s,min} = \frac{v_s}{\gamma_s} \sqrt{\frac{M v_s \omega_{s,max}}{k_B T d}} \tag{13}$$

Since s=TA/LA, the value of γ considered to be 1.8 for LA and 0.75 for TA **[9]**. M is the atom mass of graphene (2e-26 kg) and d is the ribbon width which varies from 10 to 500 nm. The temperature T under the determination of conductivity varies as a function of ribbon width and average temperature is assumed to 300K and the length is given with value 1 micrometer. $k_B$is Boltzmann's constant, ħ is the reduced Planck's constant. Vs for LA is 18,500 m/s, and for TA 16,500 m/s **[10]**

Results and Discussion

Graphene has a very high thermal conductivity compared to other carbon molecules. It has higher thermal conductivity than graphite. The thermal conductivity of graphene can be reached as high as 5000 W/m2 -k depending on the purity and width of graphene sheet.

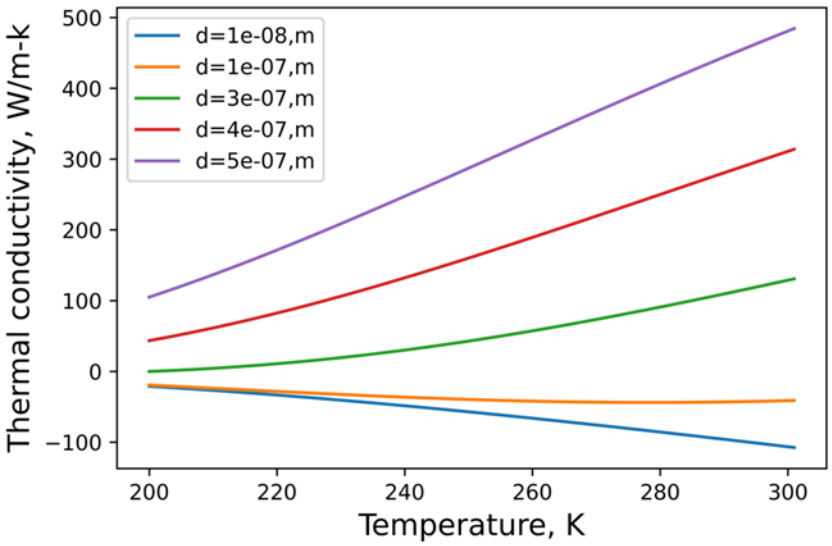


Fig 1. Thermal conductivity vs temperature for different width of graphene sheet

Thermal conductivity in nanoscale is dependent on medium's size and shape. When the mean

free path of phonon is comparable to width of the medium, the macroscale values of thermal conductivity changes. From Fig 1, we can see that with the change in temperature, thermal conductivity of graphene increases in the range of T (=200k to 300K). When temperature of a medium increases up to certain level (for low temperature), lattice vibrations also increase. As a result, number of energy carrier or phonon increases. Phonon contributes up to 99% of heat conduction in graphene.

From Fig 1, we can also see that the thermal conductivity reaches at maximum value of 500 W/m2-k. But, for the width 10 nm and 100 nm, thermal conductivity values are negative, which is wrong. This is because the widths in these cases are much lower than phonon mean free path (800 nm) for graphene. Although, others value of conductivity is positive, but it is lower compared to phonon conductivity for larger width.

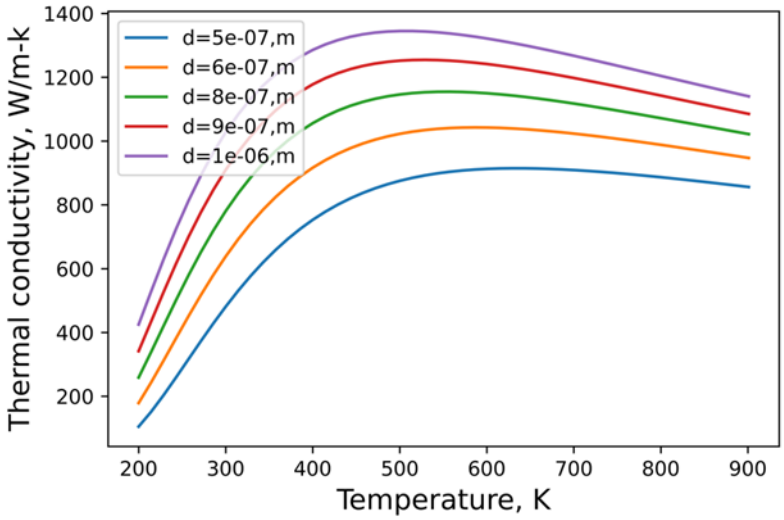


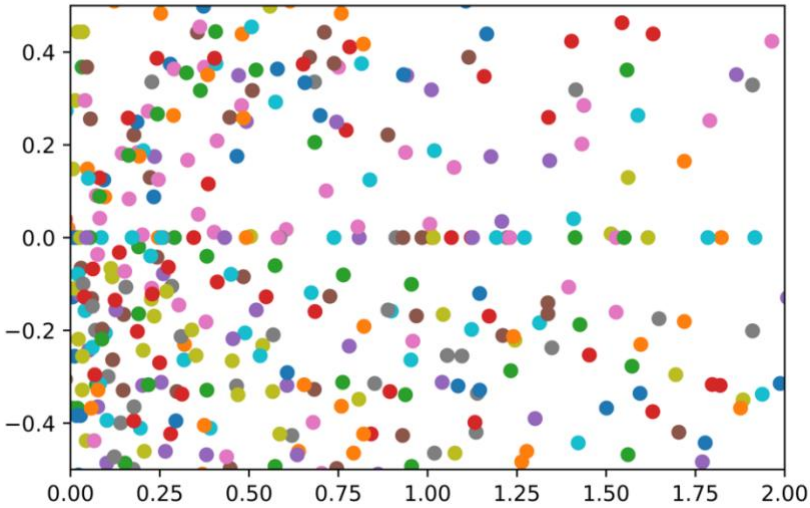
Fig 2. Thermal conductivity vs temperature

From Fig 2, we can see that thermal conductivities reach a maximum value at almost 500K for all the different widths of graphene sheet, then start to decline. Although the number of phonons and lattice vibrations increase with rising temperature, after a certain temperature due to rapid motion and collision of phonons, conductivity start to decrease.

Monte Carlo Simulation

We use Monte Carlo Simulation only to simulate the ballistic phonon distribution inside the graphene ribbon. For this simulation, we consider the system like particles in a box system. Phonons generated at (0,0) position initially, then start propagated. We consider a 2-wall graphene nanoribbon with 1 micron meter long in 2-D for this distribution, see Fig 3.

Hot Wall Top Wall (Insulated) Cold Wall



Bottom Wall (Insulated)

We initiated the phonon with random normal velocity and simulated each phonon within 10 second of time span. Details of the simulations code are shown at Appendix-1. Phonons reflected when they heat either top or bottom walls, but they are absorbed when they heat the right wall. From Fig 3, we can see the distribution of phonon with passing time.

Conclusion

Graphene has a very high thermal conductivity depending on its width and temperature. Our model of calculating thermal conductivity satisfies the results from the article **[9]**. Thermal conductivity in nanoscale is dependent on medium's size and shape. When the mean free path of phonon is comparable to width of the medium, the macroscale values of thermal conductivity changes. The thermal conductivities reach a maximum value at almost 500K for all the different widths of graphene sheet, then start to decline. Although the number of phonons and lattice vibrations increase with rising temperature, after a certain temperature due to rapid motion and collision of phonons, conductivity start to decrease. Monte Carlo Simulation is used to simulate the ballistic phonon distribution inside the graphene ribbon. We could not prepare the appropriate Monte Carlo simulation code for our project. We will try to build a better coding in future to explore the distribution of phonons and solve BTE.

Research Proposal

From our project, the equation for thermal conductivity is determined by phonon relaxation time and Umklapp scattering process. The distribution of temperature and width is calculated

and plotted by the python code. The question remains to us is about using Monte Carlo simulation to generate random phonon passing from how wall to cold wall of the graphene nanoribbon. We could not find appropriate python solution to that, which will be our goal to figure out the exactly process about how Monte Carlo simulation for the particle in the box or in a channel with top and bottom side reflecting after the phonons have collision with them.

Reference

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Appendix

```
import numpy as np
import matplotlib.pyplot as plt
import matplotlib_inline
import sympy as sym

num = 100      #initial number of phonon
num_initial  = num
xMin = 0
xMax = 2
yMin = -0.5
```

```
yMax = 0.5

# initialize positions and velocities to zero
pos = np.zeros((num,2)) # pos will represent the x,y coordinates
of the particles
vx = np.random.normal(loc = 0, scale = 1, size = (num,1))  # FILL
IN PARENTHESES!!  x-velocity of the particles
vy = np.random.normal(loc = 0, scale = 1, size = (num,1))  # FILL
IN PARENTHESES!!  y-velocity of the particles
print(num)
#print(vx,'\n',vy)
t = 0     #initial time step
time = 0.2 #each time step is 0.2time units (seconds, for
example)

while t < 10:
  t += 1     #increment time
  n = np.random.randint(0,10)     # Number of phonon generated in
each time
  print('New phonon generated = ', n)
  for particle in range(num):    #loop through each particle at
each time step
    ## Code from above can go here
    pos[particle,0] = pos[particle,0] + vx[particle]*time #update
x position
    pos[particle,1] = pos[particle,1] + vy[particle]*time #update
y position

    ############ Boundary Check ###############

    #if pos[particle,0] <= xMin:
     # num = num-1
    if pos[particle,0] >= xMax:
      num = num-1
    if pos[particle,1] < yMin:
      pos[particle,1] = yMin -(yMin - pos[particle,1])
      vy[particle] = - vy[particle]
    if pos[particle,1] > yMax:
      pos[particle,1] = yMax - (yMax - pos[particle,1])
      vy[particle] = - vy[particle]

    plt.plot(pos[particle,0], pos[particle,1], 'o') ## plot the
x-y coordinates here
    plt.xlim(0,2)
```

```
    plt.ylim(-0.5,0.5)
    plt.xlabel('')
    plt.show

print('Phonon transported to cold side from hot side = ',
num_initial - num)

from google.colab import files
plt.savefig('MAE7325_monte.png', dpi=1200, bbox_inches='tight')
files.download('MAE7325_monte.png')
```