Non-Fourier Heat Conduction of Single-Walled Carbon Nanotubes

Junichiro SHIOMI and Shigeo MARUYAMA

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1. Introduction

Non-equilibrium heat conduction can not be properly described by the Fourier's law which implies unrealistic heat propagation with infinite speed. A well known remedy for this is to view heat conduction as wave propagation described with the conventional hyperbolic energy equation [1],

$$\tau \frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} = \alpha \Delta T \,, \qquad (1)$$

where heat is conducted as a wave whose amplitude decays with an effective relaxation time τ. Here, α is the thermal diffusivity. The equation has the same asymptotic solution as the diffusion equation based on Fourier's law when $t >> \tau$, hence the smaller time/length scale of the system is, the more the difference between two solutions becomes evident.

As the new era of nanotechnology shed light to nanoscale physics, the classical problem of heat waves [2] has been revisited in last decades [3-5]. In the situations where femto or pico second heat pulse is generated by ultra pulse lasers, the finite relaxation time of the heat transport can have a significant impact on the overall physics. Although, there have been a few studies that attempted direct observations of such heat waves in crystals by means of molecular dynamics [3], due to the usually very short relaxation time of coherently excited energy in multi-dimensional systems, a clear visualization is not available to this date. In this report, we investigate a single walled carbon nanotube (SWNT) as a candidate to posses above stated properties of non-Fourier heat conduction.

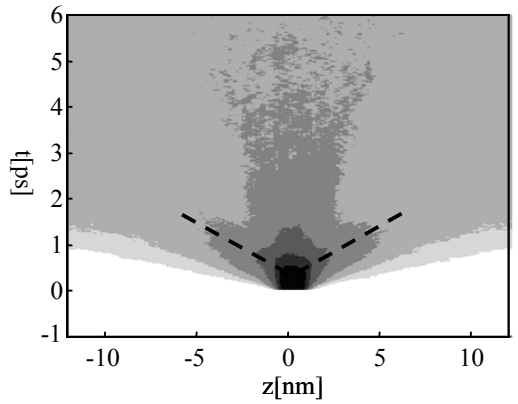

Fig. 1 Spatio-temporal isotherms of a (5,5)-SWNT subjected to a heat pulse at the origin

2. Molecular Dynamics Simulations

The molecular dynamics simulations were performed for a 25 nm long (5,5) single-walled carbon nanotube subjected to periodic boundary conditions. The carbon interactions were expressed by the Brenner potential function [6], where the total potential energy of the system is expressed as

$$E = \sum_i \sum_{j(i<j)} \left[V_R(r_{ij}) - B^*_{ij} V_A(r_{ij}) \right]. \qquad (2)$$

Here, $V_R(r)$ and $V_A(r)$ are repulsive and attractive force terms which take Morse type form with a certain cut-off function. B^*_{ij} represents the effect of the bonding condition of the atoms. As for the potential parameters, we employ the set that was shown to reproduce the force constant better (table 2 in [6]). The velocity Verlet method was adopted to integrate the equation of motion with the time step of 0.5 fs. The heat pulse was applied to a local region which consists of 6 consecutive unit cells around the center of the SWNT by connecting the region to a Nose-Hoover thermostat kept at 1000 K for time duration of 0.4 ps. The response time of the heated region to the thermostat was set to be of the order of 10fs. In order to focus on the coherent molecular motions, the total momentums of both the bulk and the heated region were cancelled. The bulk temperature of the SWNT is 50

K in order to highlight the molecular dynamics of the heat conduction at the risk of violating the quantum limit. The local temperature is defined assuming the Boltzmann distribution of the local velocity, where the statistics are acquired from 30 ensembles.

3. Results

The isotherm contours shown in Fig. 1 depict the spatio-temporal history of the temperature (z denotes the tube axial direction). The picture shows how the heat applied at the origin diffuses over the field. The heat pulse localized in several unit cells mainly excites short wavelength phonons. The minor long wavelength phonons can be still observed to form the wave fronts which propagate at the Debye group velocity of acoustic phonons (longitudinal, twisting and transverse). The interesting feature of the contour plot is the energy transport visualized as streaks stretching from near the origin (dotted lines) at the group velocity slower than the Debye group velocity of all the acoustic branches. Since the peaks of the streaks travel away from the heated region, the heat conduction is clearly non-Fourier and exhibits wave-like propagation.

In order to understand the role of each phonon mode in the above mentioned heat waves, we carried out wavelet transformations on time signals of all the carbon atoms using the Morlet wavelet as a mother wavelet. Fig. 2 shows the temporal sequences of spectrum profiles of the radial component which carries the major energy.

As seen in Fig. 2, there are two bands with distinct energy appearing around 9 THz and 18 THz. The former one has the longest life time among the visible bands. On studying the dispersion relation of the SWNT, 9 THz matches the frequency of transverse acoustic phonon modes (degenerated) in short wavelength regime. Note that the short wavelength phonons travel slower than the Debye group velocity since the dispersion saturates near the Brillouin zone boundary. The dispersion relation also shows the possible contribution to the wave-like heat conduction from some low frequency optical phonons.

The peak amplitude of the dominant energy in radial direction relaxes exponentially, which agrees with the wave equation (1). This implies that the transport of these short wavelength phonons is diffusive, instead of being ballistic, i.e. the mean free path of the phonons in this band is smaller than the characteristic length of the current system. The relaxation time τ was calculated to be 0.53 ps.

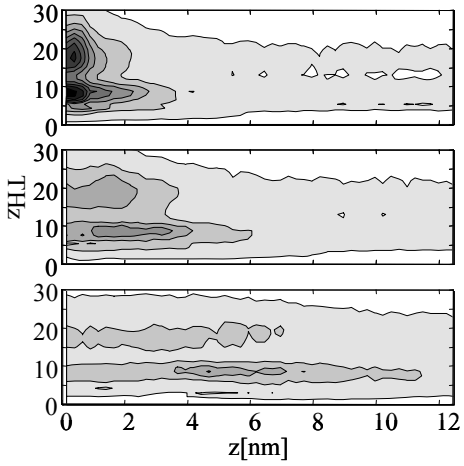

Fig. 2 Time sequence of radial spectra profile in the axial direction.

4. Conclusions

By using molecular dynamics simulations, the non-Fourier heat conduction was investigated in an SWNT subjected to a local heat pulse with time duration of sub-picoseconds. In the system with quasi-one dimensional thermal properties, we have demonstrated that the distinct heat is conducted as a wave. The wavelet transformations visualize short wavelength phonon bands and reveal the significant contribution of the transverse acoustic phonons and possible contribution of low frequency optical phonons. The heat wave relaxes exponentially which suggests the diffusive transport of the observed short wavelength phonons.

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