A MOLECULAR DYNAMICS SIMULATION OF HEAT CONDUCTION IN A CARBON NANOTUBE

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Heat conduction of finite length single walled carbon nanotubes (SWNTs) was simulated by the molecular dynamics method with the Tersoff-Brenner bond order potential[1]. Temperature at each end of a SWNT was controlled by the phantom technique, and the thermal conductivity was calculated from the measured temperature gradient and the energy budgets in phantom molecules. The thermal conductivity was measured for three different diameter SWNTs with various lengths from 6 nm through 404 nm. Since the photon mean free path is estimated to be order of 100nm ~ 1μm, heat conduction of nanotubes with shorter than 1μm should have the nearly 'ballistic' features with much less apparent thermal conductivity than infinitely long nanotubes. The Fourier's law of heat conduction may not be obeyed for these almost one-dimensional materials when rather high heat-flux conditions. The measured thermal conductivity for smaller diameter (5,5) nanotube did not converge to a finite value with increase in tube length, but an interesting power law relation was observed. The basic heat conduction mechanism was explored through the phonon dynamics extracted from the molecular dynamics simulations. The phonon density of states and photon dispersion relations were directly extracted from the simulated trajectories.

INTRODUCTION

One of purposes of this study is to clarify the thermal conductivity of carbon nanotubes, which is speculated to be higher than any other material along the cylindrical axis[2-6]. Very recently, measurements of thermal conductivity of a 5 μm thick deposited "mat" of SWNTs were reported for magnetically aligned[2] conditions. Comparing with the temperature dependence of electrical conductance in the same condition, it was concluded that the contribution of electrons to the thermal conductivity is negligible in all temperature range. The direct measurements of individual nanotube were also tried with the MEMS assisted new measurement technology[3]. Quickly following those experiments, several preliminary molecular dynamics simulations[4-6] showed very high thermal conductivity such as 6600 W/mK at 300 K[4]. However, the estimated values of thermal conductivity were widely different from one another. Another purpose of this study is the preliminary connection of molecular dynamics techniques to the solid-state heat conduction usually discussed as "phonon transport" in solid physics. In principle, the molecular dynamics simulation should be used to obtain information for phonon transport dynamics such as phonon dispersion relation, group velocity, mean free path, boundary scattering rate and the rate of phonon-phonon scattering (Umklapp process). It is also anticipated that by developing the phonon concept to more general form in order to understand the thermal boundary resistance even in the liquid-solid interface.

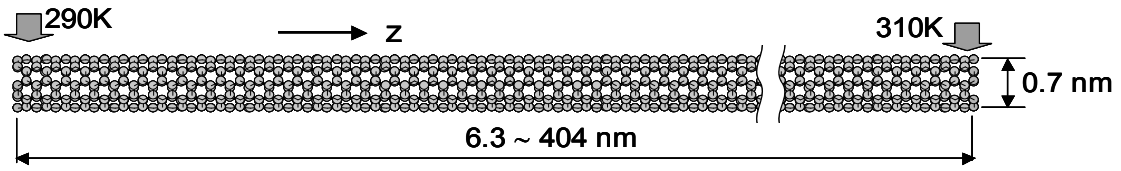

Fig. 1 Simulation system for (5,5) natotube.

SIMULATION TECHNIQUE

Three armchair type SWNT structures with different diameters (5,5), (8,8) and (10,10) were chosen. Here, (10,10) has been most widely studied structure of SWNT. On the other hand, (5,5) has the almost similar diameter as C_{60} and the inexpensive and huge scale production of SWNTs with this diameter is anticipated with the new generation technique using high-pressure and high-temperature CO gas. By applying the phantom heat bath model to each end of a SWNT, the temperature difference was applied. Here, no periodic boundary condition was applied to handle the finite size effect of carbon nanotubes. It is often discussed that the cell length of periodic boundary condition should be larger than the "mean free path" of phonon, which is argued to be order of 1 μm though it is really arbitral value depending on the definition. The length of SWNTs was varied from 6 nm through 404 nm for (5,5) nanotube. With our configuration, thermal conductivity was calculated from the measured temperature gradient and the heat flux obtained by the energy budgets of phantom molecules.

RESULTS

Thermal Conductivity

An example of the temperature distribution along the tube is shown in Fig. 2 for (5,5) and 202nm case (16000 atoms). The phantom temperatures at each end were set as 290 K and 310 K. After obtaining the temperature about 300 K with the auxiliary velocity scaling temperature control, typically, 1 ns simulations were performed for the equilibration with only phantom temperature control. Then, 1 ~ 2 ns calculation was used for the measurement of temperature distribution. With the controlling energy budgets of phantom molecules, the heat flux along the tube can be simply calculated. Combined with the temperature gradient such as in Fig. 2, the thermal conductivity λ can be calculated through Fourier's equation: $q = -\lambda(\partial T / \partial y)$. As the cross-sectional area, a ring of van der Waals thickness 3.4 Å was used. The dependence of the thermal conductivity on the nanotube length is summarized in Fig. 3. The thermal conductivity was diverging with the power-law characteristics with nanotube length at least up to 0.4 μm long nanotube. This very striking behavior of thermal conductivity for especially (5,5) is similar to the one-dimensional

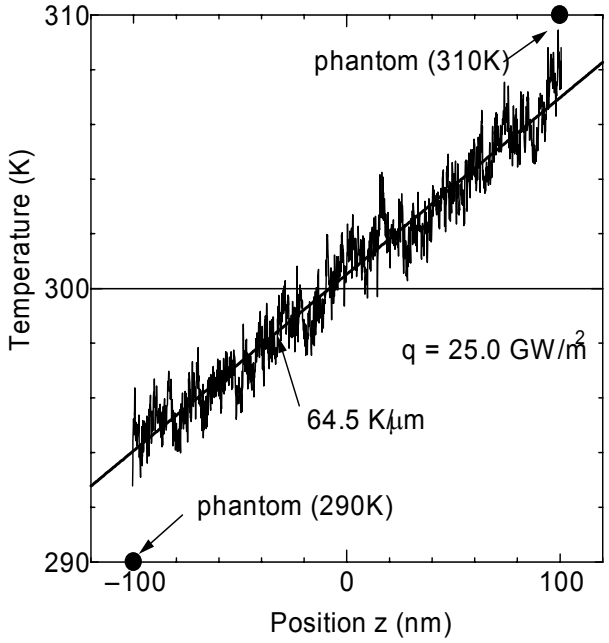

Fig. 2. Temperature distribution along a (5,5) nanotube with 202nm length.

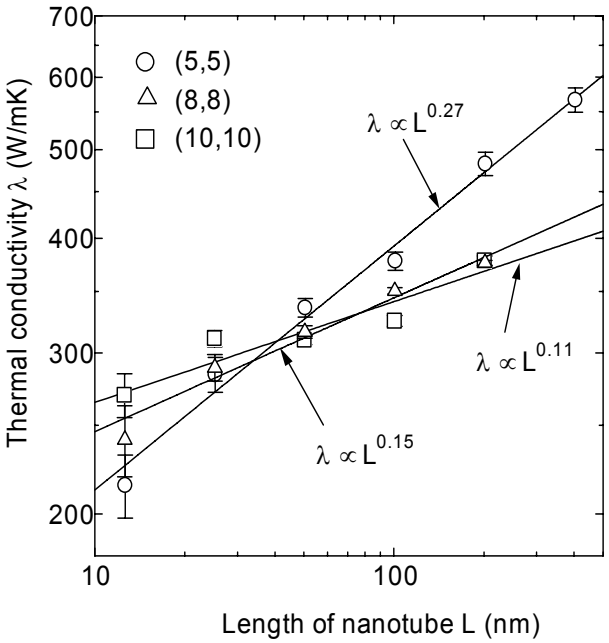

Fig. 3. Dependence of thermal conductivity on length of nanotubes for 300K.

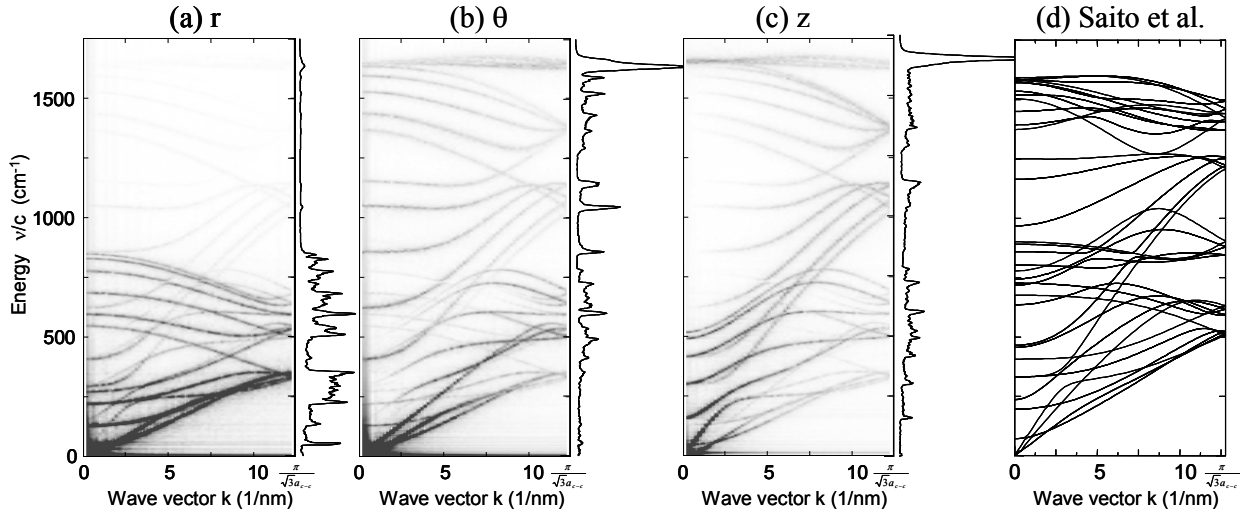

Fig. 4 Phonon dispersion relation and photon density of states for (5,5) SWNT.

model calculations of thermal conductivity[7] where the divergence of λ with the power of 0.35 or 0.4 is discussed. However, (5,5) nanotube is real physical material. The thermal conductivity may converge when the tube length is much longer than the mean free path of energy carrying phonon. However, the thermal conductivity of about 600 W/mK for about 0.4 μm tube is still increasing with the power law.

Phonon Dispersion Relation

The basic heat conduction mechanism was explored through the phonon dynamics extracted from the molecular dynamics simulations. The phonon density of states was measured as the power spectra of velocity fluctuations in eq. (1) as in the right-hand side insert in Fig. 4.

$$D_\alpha(\omega) = \int dt \exp(-i\omega t)\langle v_\alpha(t) v_\alpha(0)\rangle \quad (1)$$

The photon dispersion relations were also directly measured as the time-space 2-D Fourier transforms in eq. (2) of the position of each molecule as in Fig. 4.

$$R_\alpha'(k,\omega) = \int dt\, r_\alpha'(z,t) \exp(ikz - i\omega t) \quad (2)$$

The theoretical 36 lines of phonon dispersions were reproduced as in the previous theoretical results[8]. The group velocities of four acoustic phonons were estimated as longitudinal acoustic mode: 17 km/s (z-direction), transverse acoustic mode (degenerate): 7 km/s (r-direction), twisting acoustic mode: 10 km/s (θ-direction), in good agreement with the previous estimations[8].

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