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A Molecular Dynamics Simulation of Heat Conduction in Finite Length SWNTs

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Abstract

The heat conduction in finite length single walled carbon nanotubes (SWNTs) was simulated by the molecular dynamics method with the Tersoff-Brenner bond order potential. 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 (5,5) and (10,10) SWNTs with various lengths from 6 nm through 404 nm. Measured thermal conductivity for smaller diameter (5,5) nanotube did not converge to a finite value with increase in tube length, but obeyed a striking power law relation. The phonon density of states and phonon dispersion relations were calculated from the simulated results for the better understanding of the heat conduction mechanism.

Keywords: Thermal Conductivity, Molecular Dynamics Method, Phonon, SWNT

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Introduction

The thermal conductivity of single walled carbon nanotubes (SWNTs) is speculated to be higher than any other materials along the cylindrical axis. Recently, measurements of thermal conductivity of 5 μm thick magnetically aligned deposited 'mat' of SWNTs were reported [1]. Comparing with the temperature dependence of electrical conductance in the same condition, it was concluded that the contribution of electrons to the thermal conductivity was negligible in all temperature range. Besides those experiments, several molecular dynamics simulations showed very high thermal conductivity such as 6600 W/mK at 300 K [2]. Since the phonon mean free path is estimated to be order of 100 nm ~ 1 μm, thermal conductivity of nanotubes shorter than a few μm should have the nearly 'ballistic' features with much less apparent thermal conductivity compared to the infinitely long nanotubes. The finite length effect on the heat conduction is explored in this paper.

Simulation Techniques

Using the Tersoff-Brenner bond order potential [3], 2 armchair types SWNT structures with different diameters, (5,5) and (10,10), were simulated by the molecular dynamics method. Here, (5,5) has the almost similar diameter as C_{60} and the huge scale production of SWNTs with this diameter is expected with the new generation technique using high-pressure and high-temperature CO. Here, no periodic boundary condition was applied in order to explore the finite size effect of carbon nanotubes. The length of SWNTs was varied from 6 nm through 404 nm for (5,5) nanotube. By applying the phantom heat bath model to each end of a SWNT, temperature difference was applied. For the average temperature of 300K, phantom temperatures at each end were set as 290 K and 310 K. Typically about 1 ns simulations was necessary for the equilibration with phantom temperature control, and 1 ~ 2 ns calculation was used for the measurement of temperature gradient.

Thermal conductivity λ was calculated by Fourier's equation $q = -\lambda(\partial T / \partial z)$ with the measured temperature gradient $\partial T / \partial z$ and the heat flux q calculated from the energy budgets of phantom molecules. As the cross-sectional area, a ring area of van der Waals thickness 3.4 Å was employed.

Results and Discussions

The dependence of the thermal conductivity on the nanotube length is summarized in Fig. 1. Even though the thermal conductivity was almost constant for (10,10) independent of the tube length, it was diverging for smaller diameter (5,5) case. This striking behavior of thermal conductivity for (5,5) is similar to the one-dimensional model calculations of thermal conductivity [4] 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 is still increasing with the power law up to about 0.4 μm nanotube. Because the divergence for (10,10) is not apparent or very weak in Fig. 1, it is speculated that the reason for the divergence is not simply the short length compared to the phonon mean-free-path but the limited freedom of motion for thin (5,5) nanotube.

The 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 as shown in the right-hand side of Fig. 2. The phonon dispersion relations in Fig. 2 were also directly measured as the time-space 2-D Fourier transforms of the position of each molecule as $R'(k,\omega) = \int dt r'(z,t) \exp(ikz - i\omega t)$. The theoretical 36 lines of phonon dispersions were reproduced in Fig. 2. The group velocities of four acoustic phonons were estimated as 17 km/s (longitudinal acoustic mode clearly seen in z-projection), 7 km/s (transverse acoustic mode in r-projection), and 10 km/s (twisting acoustic mode in θ-projection), in good agreement with the previous estimations [5].

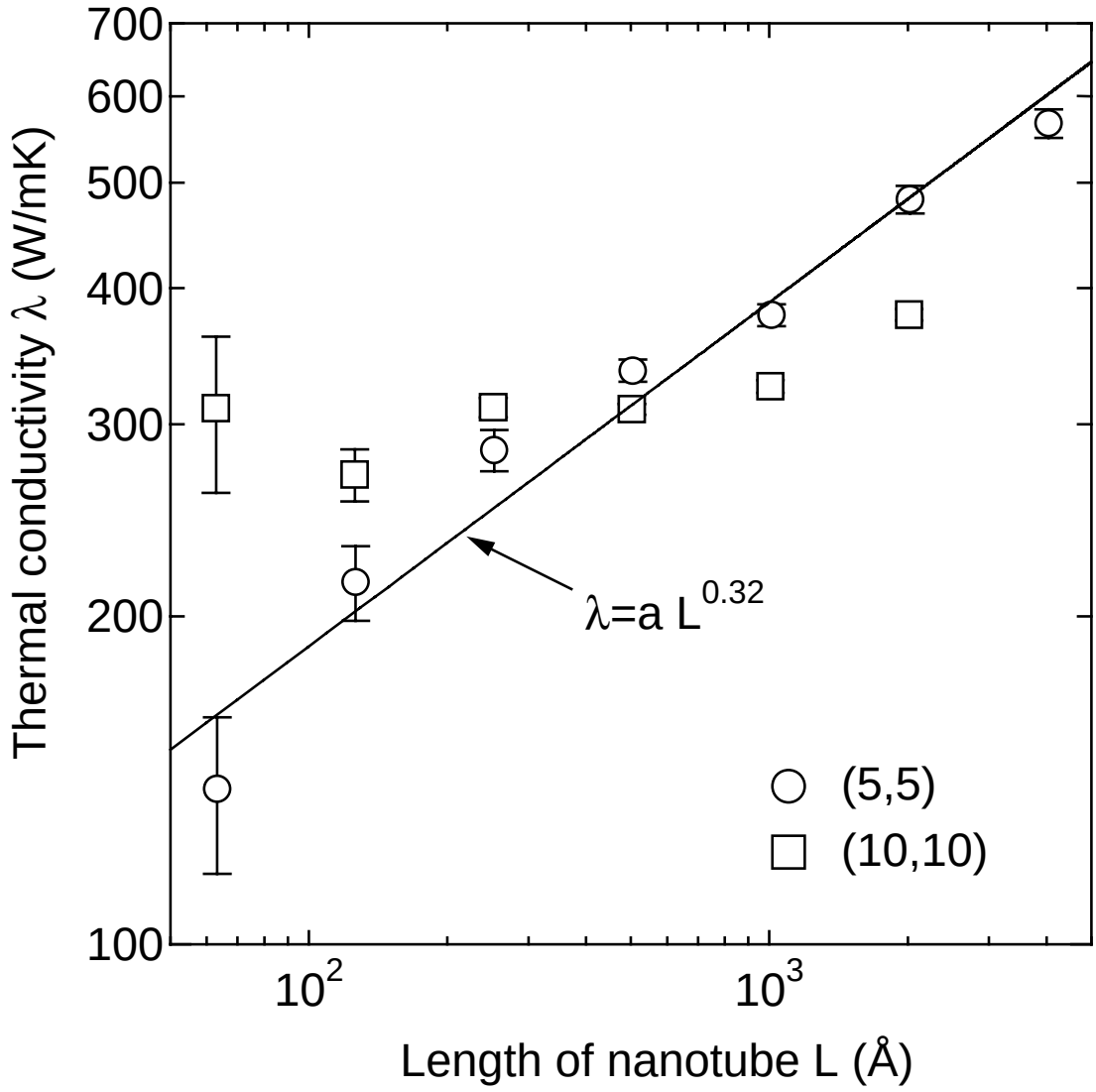
References

1. J. Hone, et al., Appl. Phys. Lett., 77 (2000), 666.
2. S. Berber, et al., Phys. Rev. Lett., 84-20 (2000), 4613.
3. D. W. Brenner, Phys. Rev. B, 42-15 (1990), 9458.
4. S. Lepri, Eur. Phys. J. B, 18 (2000), 441.
5. M. S. Dresselhaus, and P. C. Eklund, Adv. Phys., 49-6 (2000), 70.

Captions to Figures

Fig. 1. Dependence of thermal conductivity on length of nanotubes for 300K.

Fig. 2. Phonon dispersion relation and phonon density of states for (5,5) SWNT.



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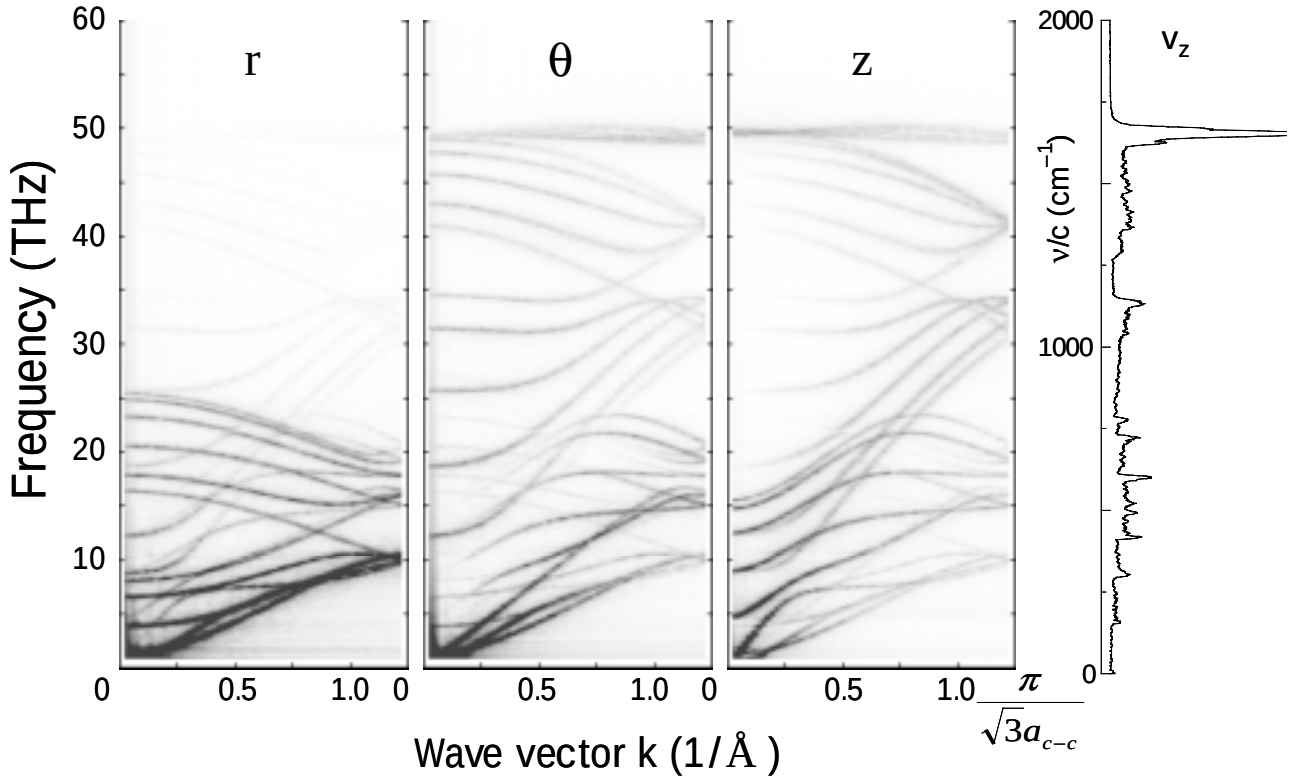

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