Heat conduction in disordered semiconductor carbon nanotubes

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High thermal conductivity is required for high performance electronics and one advantage of carbon nanomaterials as candidates for future electronics is that they have peculiar thermal conductivites. In this letter, we address the effects of Anderson type disorder to lattice thermal conductivity through long carbon nanotubes (CNTs) using atomistic Green functions. The dependence of phonon transmission is analyzed as a function of the length of the disordered CNT, thermal conductance as a function of temperature is calculated for different lengths. Analysis of transmission amplitudes as a function of length of the disordered device shows that phonons with different energies display different transport regimes, i.e. quasi-ballistic, diffusive and localization regimes coexist. In the light of the results we discuss heating of the semiconductor CNT device in electronic applications.

INTRODUCTION

Thermal transport in low-dimensional systems is of central importance for applications. By properly controlling thermal properties it is possible to enhance the device performance. In electronic applications, high values of both electronic and phononic conductances are desired. On the other hand a low phonon conductance is required in order to have efficient thermoelectric energy conversion. In semiconductor materials heat transport is governed mostly by phonons and it is previously shown that the thermal conduction is strongly influenced by device dimensions [1]. For example, it was observed recently that phonon conduction can be reduced without effecting the electronic transport importantly in Si nanowires which leads to an enhancement in the thermoelectric figure of merit by two orders of magnitude compared to its bulk value [2, 3, 4]. This decrease in thermal conduction is mainly due to the scattering of phonons at disordered surface whereas electron transfer is maintained by bulk-like states. Thus, surface to volume ratio is a parameter to control the transport properties of these devices. Graphitic allotropes have the exceptional property that they are one atom tick. There is a growing interest in the field of phononic energy transport through carbon based materials. It is shown that thermal conductance of nanotubes can be tuned by sliding the inner shell inside the outer shell [5]. Disorder induced localization is believed to be a possible explanation of the exponential dependence of thermal resistance on the telescoping distance. Conductance is independent of the device length if the device is pristine. Real systems, on the other hand, always include disorder which we model with a distribution in force constants. Recently the effect of isotopic disorder on thermal conduction through nanotubes is studied theoretically [6] and good agreement with experiment [7] is achieved.

In this letter, we analyze the effects of disorder on thermal transport through semiconductor CNTs. First we summarize the atomistic Green function method in calculating the transport properties. Then we investigate the dependence on length of the device and on the operating temperature. The energy dependence of transport regimes are shown to coexist and their role on device performance depending on system parameters is discussed.

METHOD

As it is common in transport calculations, we apply the partitioning scheme and divide the system as left, center and right regions (L, C, and R respectively). In the harmonic approximation the vibrational Hamiltonian can be written as

$$H = \sum_{\alpha} H_{\alpha} + \left(u^{L}|K^{LC}|u^{C}\right) + \left(u^{R}|K^{RC}|u^{C}\right) \qquad (1)$$

where the first term $H_{\alpha} = 1/2\,(\dot{u}^{\alpha}|\dot{u}^{\alpha}) + 1/2\,(u^{\alpha}|K^{\alpha\alpha}|u^{\alpha})$ is the Hamiltonian of the decoupled subsystem α, the second and third terms stand for the coupling between the central region and the reservoirs. Here $|u^{\alpha})$ is the vector, and $(u^{\alpha}|$ is its transpose, of mass renormalized coordinates $u_i^{\alpha} = \sqrt{m_i} x_i^{\alpha}$, x_i^{α} being the i^{th} degree of freedom of subsystem α with m_i the mass associated to this degree of freedom. $K_{ij}^{\alpha\beta}$ is the matrix element representing the coupling between mass renormalized coordinate i of subsystem α with j subsystem β, and $K_{ij}^{\alpha\beta} = k_{ij}^{\alpha\beta}/\sqrt{m_i m_j}$ with $k_{ij}^{\alpha\beta}$ being the spring constant in direct coordinates. In our calculations, we make use of force constants with fourth nearest neighbor parameters which are shown to have good agreement with density functional theory calculations [8] [see Fig. 1].

We calculate the phonon transmission using atomistic Green functions. Retarded Green function for a subsystem in the absence of coupling to the reservoirs is defined with $g^{r(a),\alpha}[\omega] = \left[(\omega \mp \mathrm{i}0^{+})^2 I - K^{\alpha\alpha}\right]^{-1}$ with I being the identity matrix. Self energy due to coupling to reservoir α is $\Sigma^{r(a),\alpha} = K^{C\alpha} g^{r(a),\alpha} K^{\alpha C}$ and the retarded function for the central region in the presence of

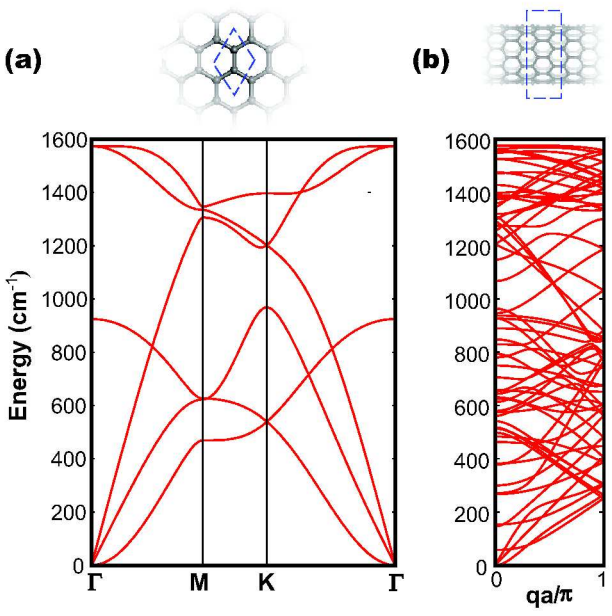

FIG. 1: Phonon dispersions of graphene (a) and CNT(10,0) (b). Lattice structures are shown in the upper part; unit cells are indicated with dashed lines.

reservoirs reads

$$G^r[\omega] = \left[(\omega + \mathrm{i}0^+)^2 I - K^{CC} - \Sigma^{r,L} - \Sigma^{r,R}\right]^{-1}. \quad (2)$$

In our calculations, we calculate the free Green function of the semi-infinite reservoirs using the surface Green function matching method [9] and the broadenings with the relation $\Gamma^\alpha = \mathrm{i}\left(\Sigma^{r,\alpha} - \Sigma^{a,\alpha}\right)$. Thermal conductance is defined as

$$\kappa(T) = \int_0^\infty \frac{d\omega}{2\pi}\,\hbar\omega \frac{\partial f_B(\omega,T)}{\partial T}\mathcal{T}(\omega) \quad (3)$$

where $\mathcal{T}(\omega) = \mathrm{Tr}\left[G^r\Gamma^L G^a \Gamma^R\right]$ is the transmission function, f_B is the Bose distribution with ω being the angular frequency and T being the temperature. We introduce Anderson type disorder by a random distribution of m_i at the C region which transforms to a distribution of disordered coupling matrix elements K_{ij}^{CC}. Green functions of the disordered region are obtained using decimation techniques and the transmission functions are obtained by averaging over 250 sample configurations.

RESULTS AND DISCUSSIONS:

First of all we note that, unlike its electronic counterpart, phononic heat transmission takes place not only in a small energy window but phonons of all energy values contribute to the conductance. In this sense, low energy phonons play a special role in phonon thermal conduction in both pristine and disordered systems as it will be

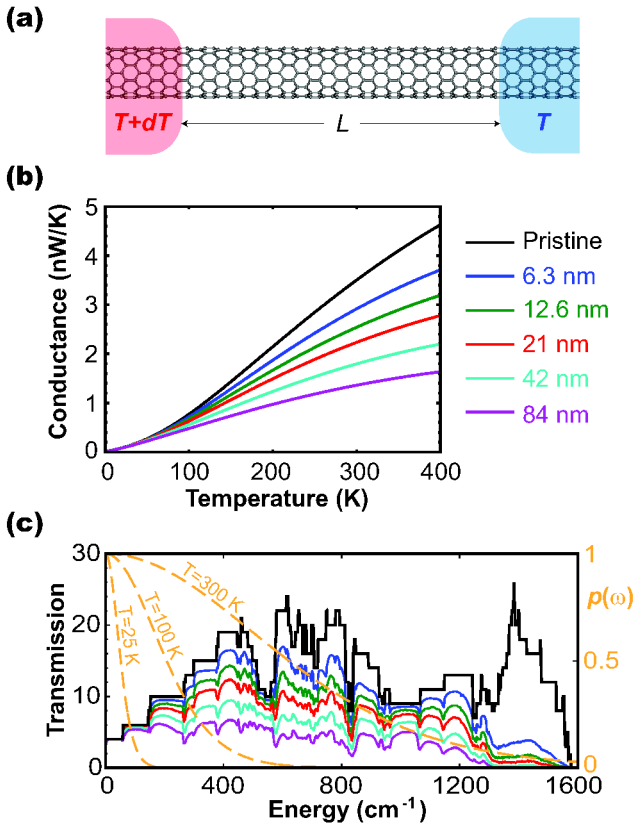

FIG. 2: (a) Schematics of the system under consideration. (10,0) CNT is placed between reservoirs which have a temperature difference dT. We include Anderson type disorder at the central region whose length L is varied. (b) Thermal conductance as a function of temperature T for (10,0) CNTs with diffrent lengths. (c) Transmission versus energy for CNTs with diffrent lengths L (solid curves). Black curve is the transmission function of pristine (10,0) CNT. Dashed curves are the prefactors $p(\omega,T)$ for T =25, 100, 300 K (see text). (Same color code is used in subplots (b) and (c)).

discussed below. We consider (10,0) CNT which is placed between reservoirs made of the same material [Fig. 2(a)]. The reservoirs are kept at different temperatures T and $T + dT$. We introduce Anderson type disorder in the central region and analyze the dependence of the transmission function and thermal conductance to disorder for CNTs with different lengths. The transmission function for the pristine CNT reflects the fact that each phonon branch is contributing to transport with unit probability. Namely, the transmission function at energy ω is the number of phonon branches $N(\omega)$ crossing at this energy each acting as an independent transmission channel [Fig. 1(b)]. Increasing the length of the C-region, the transmission is reduced but the low energy transmission is almost unaffected by disorder [Fig. 2(c)]. On the other hand high energy phonons are extremely sensitive to disorder in the sense that the transmission is blocked

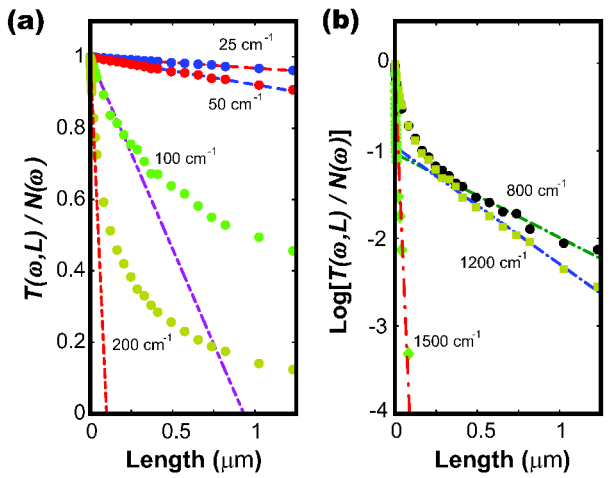

FIG. 3: Different regimes of phonon transport through (10,0) CNTs. Normalized transmission values are plotted as a function of length for various energies. At low energies (25 cm^{-1}, 50 cm^{-1}) $\mathcal{T}(\omega)/N(\omega)$ can be fit to a line (a). Intermediate energies like 100 cm^{-1} and 200 cm^{-1} deviate from the linear behavior and show $1/L$ behavior (a). High energy phonons are best described with an exponential law (b).

to a great extend even for the shortest regions with disorder. We also note that disorder induced scattering is more important near van Hove singularities in the density of states. We observe that thermal conductance drops as the length of the central region is increased but this drop is significant only at high temperatures [Fig. 2(b)]. As the temperature increases the difference in conductance for different lengths increase and then saturates. The low temperature thermal conductance is insensitive to disorder because of two reasons. First, because the transmission of low energy phonons itself is insensitive to disorder. Second, the term $p(\omega,T) = \omega\,\partial f_B(\omega,T)/\partial T$ in the integrand of Eqn. 3 is filtering out the contribution of high energy phonons to low temperature conductance. At high temperatures $p(\omega)$ flattens and the filter effect diminishes.

In order to identify the different transport regimes, we analyze $\mathcal{T}(\omega,L)/N(\omega)$ for a number of ω values, and as a function of L. Transmission amplitudes are normalized in this way using their pristine values $N(\omega)$ in order to enable comparison. In Fig.3(a), it is shown that low energy transmission (ω =25, 50 cm^{-1}) decreases linearly with L reflecting the fact that low energy phonons display a quasi-ballistic behavior. For higher energies (ω =100, 200 cm^{-1}) the transmission function deviates from the linear fit to short device transmission (see Fig. 3(a)) and a diffusive behavior is observed. On the other hand, energetic phonons (ω =800, 1200, 1500 cm^{-1}) experience localization at different lengths depending on their energies as it is shown in Fig. 3(b). Different transport regimes coexist for phonons with different energies. Altough high energy phonons experience localization, a pure localization regime or an exponential dependence of thermal conductance on CNT length is not observed.

Therefore heating of the semiconductor CNT device in electronic applications is dependent on the length of the device and the frequency distribution of generated phonons. Low energy phonons will be discharged effectively while localization of energetic phonons may cause overheating of long devices. Localization induced heating becomes more dramatic at high operating temperatures.

CONCLUSIONS

In summary, we analyzed the effects of disorder on phonon thermal transport of semiconductor CNTs. We show that different transport regimes for phonons of different energies coexist and their relative weight on thermal conductance depends on temperature. Long wavelength phonons are transmitted effectively but short wavelength phonons are likely cause overheating and the effect localization will be more pronounced at higher operating temperatures.

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[1] M. S. Dresselhaus, G. Dresselhaus, M. Hofmann, J. Vac. Sci. Technol. B **26**, 1613 (2008).
[2] A. I. Hochbaum *et. al*, Nature **451**, 163 (2008).
[3] A. I. Boukai *et. al*, Nature **451**, 168 (2008).
[4] T. Markussen, A.-P. Jauho, M. Brandbyge, Phys. Rev. B **79**, 035415 (2009).
[5] C. W. Wang *et. al*, App. Phys. Lett. **90** 193114 (2007).
[6] I. Savić, N. Mingo, D. A. Stewart, Phys. Rev. Lett. **101**, 165502 (2008); D. A. Stewart, I. Savić, N. Mingo, NanoLett. **9**, 81 (2009).
[7] C. W. Chang, A. M. Fennimore, A. Afanasiev, D. Okawa, T. Ikuno, H. Garcia, Deyu Li, A. Majumdar, A. Zettl, Phys. Rev. Lett. **97**, 085901 (2006).
[8] J. Zimmermann, P. Pavone, and G. Cuniberti, Phys. Rev. B **78**, 045410 (2008).
[9] M. P. Lopez-Sancho, J.M. Lopez-Sancho, and J. Rubio, J. Phys. F **14**, 1205 (1984).