Computational Materials Science 103 (2015) 151–156

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Phonon–phonon scattering rates in single walled carbon nanotubes

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ARTICLE INFO

Article history:
Received 16 January 2015
Accepted 23 February 2015
Available online 8 April 2015

Keywords:
Phonon–phonon interaction
Scattering rate
Single walled carbon nanotube

ABSTRACT

A theoretical model for the phonon–phonon scattering rates of carbon nanotubes (CNTs) is developed using the carbon specific Brenner's potential. This model allows for the calculation of mode specific phonon–phonon scattering rates, via direct computation of the three-phonon strength of interaction. This direct calculation provides further accuracy to the previously existing model, which relied on continuum mechanics approximations to the strength of interactions. The contributions of each phonon branch to the total phonon–phonon scattering rates are analyzed. The results for longitudinal optical and longitudinal acoustic phonons of a (10, 10) CNT suggest very different behavior for each vibrational mode. While the results presented are specific to the (10, 10) metallic CNTs, the method is directly applicable to CNTs of other chiralities.

1. Introduction

It is very difficult to obtain Carbon Nano Tube (CNT) material properties for specific chiralities experimentally,as a result computational modeling has been extensively used to obtain thermal, mechanical, electrical properties [1–13]. In order to model and compute electrical conductivity and Joule heating, it is necessary to calculate electron–phonon scattering rates in a CNT, using models proposed in the literature [2–5,14,15].

For metallic CNTs under electrical loading, the continued scattering between electrons and phonons results in a non-equilibrium phonon distribution associated with the creation of hot phonons. The effects of hot phonons on Joule heating [16] and wind forces [17] have been reported. If these hot phonons are not allowed to relax back toward the equilibrium phonon distribution, the creation of phonons in electron–phonon scattering process may result in a net accumulation of hot phonons. To avoid such pile-ups of phonons during the simulation of CNTs, it is important to understand and calculate the strength of interactions between all phonons.

Several groups have studied phonon–phonon scattering rates either experimentally [18,19] or theoretically [20–24]. Raman scattering experimental results presented in Refs. [18,19] report a decay rate of optical phonons at 300 K of 1 ps and 1.1 ps respectively. In Ref. [20] Pennington and his group make use of the phonon Boltzmann transport equation to calculate the optical phonon relaxation times for various CNTs. The results presented in Ref. [20] vary from 0.2 to 10.2 ps at 300 K depending on the chirality of the CNT. While experimental and theoretical results seem to agree on the magnitude of the phonon–phonon scattering rates, they may not necessarily reflect the true behavior of each individual phonon–phonon interactions because of the averaging process used. The Raman scattering results presented in Ref. [18] are for very specific frequencies and do not show the full frequency spectrum that phonons can span. Moreover, most theoretical models for phonon–phonon scattering rates calculate constant scattering rates for all phonons. However, during three phonon processes, there is no physical reason why the scattering rates should be constant for all phonon–phonon scattering mechanisms. The use of constant rates is a very rough approximation, which can be improved by analyzing the full phonon bands. According to the ab initio study of thermal transport properties, Broido et al. [23] reported that the three-phonon scattering is the dominant phonon scattering mechanisms around room temperature. In Ref. [21], Hepplestone and Srivastava derived a formalism to calculate phonon specific scattering rates based on the anharmonic part of a generalized three dimensional lattice potential. The authors of Ref. [21] were able to show that phonon–phonon scattering rates have large variations for different phonon wavevector values. Their approach relied on the calculation of anharmonic scattering rates making use of a continuum mechanics approach to obtain the constant used in the strength of interaction formalism.

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http://dx.doi.org/10.1016/j.commatsci.2015.02.046
0927-0256/

In this study, improved accuracy is provided for the model proposed in Ref. [21] by using the carbon specific Brenner's potential [25] to directly compute the strength of interaction between specific phonons and determine the phonon–phonon scattering rates in a (10,10) CNT without the need for continuum mechanics assumptions and approximations of the strength of interaction.

2. Methodology

The analytical formula for the phonon–phonon scattering rates is derived from the interatomic bonding potential v. Performing a Taylor expansion of the potential, the harmonic and anharmonic parts of the potential can be extracted.

$$v = v_0 + \sum_{lb}\sum_{\alpha}\left.\frac{\partial v}{\partial u_\alpha(lb)}\right|_0 u_\alpha(lb) + \frac{1}{2}\sum_{lb,l'b'}\sum_{\alpha,\beta}\left.\frac{\partial^2 v}{\partial u_\alpha(lb)\partial u_\beta(l'b')}\right|_0 u_\alpha(lb)u_\beta(l'b') + \frac{1}{3!}\sum_{lb,l'b',l''b''}\sum_{\alpha,\beta,\gamma}\left.\frac{\partial^3 v}{\partial u_\alpha(lb)\partial u_\beta(l'b')\partial u_\gamma(l''b'')}\right|_0 u_\alpha(lb)u_\beta(l'b')u_\gamma(l''b'') \quad (1)$$

In Eq. (1), v_0 is the constant part of the potential, and $u(lb)$ is the displacement of an atom from its equilibrium position in a unit cell of the lattice. The vector l represents the location of the unit cell within the lattice, while the vector b points to the position of a given atom within the unit cell. α, β, and γ represent the summation over the x, y, and z coordinates. The third order term of the Taylor expansion is responsible for the anharmonic behavior of the potential, and it represents the strength of interaction in three phonons processes shown on Fig. 1.

From Eq. (1), we define the third rank tensor, Ψ, as the third derivative of the potential with respect to the displacements around the atomistic equilibrium positions.

$$\Psi_{\alpha\beta\gamma}(lb,l'b',l''b'') = \frac{\partial v^3}{\partial u_\alpha(lb)\partial u_\beta(l'b')\partial u_\gamma(l''b'')}|_0, \quad (2)$$

The details of the calculation of this tensor are presented in Section 3. Following Srivastavas approach, [26] the anharmonic bonding potential is given as,

$$v_3 = \frac{1}{3!}\frac{i}{(N_0\Omega)^{\frac{3}{2}}}\sum_{\substack{qb,q'b',q''b''\\ s,s',s'',\alpha,\beta,\gamma}}\left(\frac{\hbar^3}{8m_c^3\omega(qs)\omega(q's')\omega(q''s'')}\right)^{\frac{1}{2}} * e_\alpha(b|qs)e_\beta(b'|q's')e_\gamma(b''|q''s'') * \Psi_{\alpha\beta\gamma}(qb,q'b',q''b'') * \left(a^\dagger_{qs}-a_{-qs}\right)(a^\dagger_{q's'}-a_{-q's'})(a^\dagger_{q''s''}-a_{-q''s''})\delta_{G,q+q'+q''}, \quad (3)$$

where $N_0\Omega$ is the volume of the lattice, m_c is the mass of one carbon atom, $\omega(qs)$ is the frequency of a phonon with wavevector q and polarization s, G is a reciprocal lattice vector, $e_\alpha(b\mid qs)$, $e_\beta(b'\mid q's')$, and $e_\gamma(b''\mid q''s'')$ are the direction of the polarization vectors, $\Psi_{\alpha\beta\gamma}(qb,q'b',q''b'')$ is the Fourier transform of the third rank tensor presented in Eq. (2), finally, $a^\dagger$ and a represent the phonon creation and annihilation operators respectively. This anharmonic potential is then used to calculate the transition probabilities of three phonon processes. Before the final formula for the phonon–phonon scattering rates is derived, it is important to discuss the nature of three phonons processes. These processes can be divided into two classes. Class I represents the annihilation of two phonons and the creation of a third one, while class II represents the annihilation of one phonon and the creation of two phonons as shown on Fig. 1. Phonon–phonon scattering must satisfy the conservation of energy and momentum laws, which can be written as the following selection rules presented in Table 1.

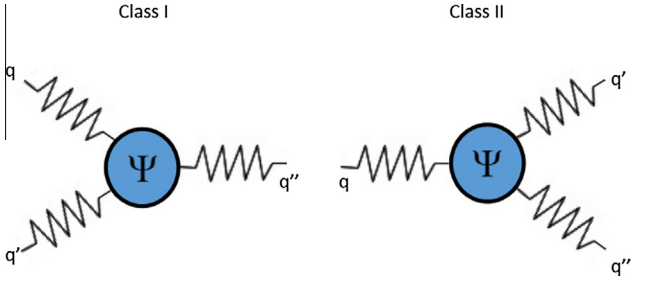

Fig. 1. Three phonon processes with interaction strength Ψ.

Table 1
Class I and class II selection rules.

Class I	Class II
$q + q' - q'' = G$	$q - q' - q'' = G$
$E(q) + E(q') = E(q'')$	$E(q) = E(q') + E(q'')$

The transition probability between an initial state $\mid i\rangle$ and a final state$\mid f\rangle$ is given by Fermi's golden rule,

$$P = \frac{2\pi}{\hbar}|\langle f|v_3|i\rangle|^2\rho, \quad (4)$$

where ρ is the density of final states. Using the following initial and final states for class I and class II processes respectively,

$$\begin{aligned} |i_{I,II}\rangle &= |n_{qs}, n_{q's'}, n_{q''s''}\rangle \\ |f_I\rangle &= |n_{qs}-1, n_{q's'}-1, n_{q''s''}+1\rangle \\ |f_{II}\rangle &= |n_{qs}-1, n_{q's'}+1, n_{q''s''}+1\rangle \end{aligned} \quad (5)$$

where n is the phonon occupation number, we can obtain the final formulation for the transition probabilities as follows,

$$P_I = \frac{\pi\hbar^2\Phi^2 n_{qs}n_{q's'}(n_{q''s''}+1)\rho\delta_{G,q+q'-q''}}{144 * m_c^3(N_o\Omega)^3\omega(qs)\omega(q's')\omega(q''s'')} \quad (6)$$

and

$$P_{II} = \frac{\pi\hbar^2\Phi^2 n_{qs}(n_{q's'}+1)(n_{q''s''}+1)\rho\delta_{G,q-q'-q''}}{144 * m_c^3(N_o\Omega)^3\omega(qs)\omega(q's')\omega(q''s'')}, \quad (7)$$

where Φ is the projection of the third rank tensor described in Eq. (2) along the directions of the polarization vectors.

It maybe expected that there should be a symmetry between the scattering rates of the two types of three phonon processes. However, we should remember that phonons are just quasi-particles, quantizations of the lattice vibration. Depending on the magnitude of vibration, the number of phonons in each mode is different. Therefore, the scattering rates of class I and class II are different, as indicated by $n_{qs}n_{q's'}(n_{q''s''}+1)$ in Eq. (6) and by $n_{qs}(n_{q's'}+1)(n_{q''s''}+1)$ in Eq. (7). Therefore, there is no symmetry between the scattering rates of class I and class II.

Making use of the relaxation time approximation, we can write the scattering rate of a phonon (qs) as,

$$\frac{1}{\tau_{qs}} = \frac{\pi\hbar^2}{144 * m_c^3(N_o\Omega)^3\omega(qs)(n_{qs}+1)} \times \sum_{q's',q''s''}\left[\frac{\Phi^2 n_{q's'}(n_{q''s''}+1)\rho\delta_{G,q+q'-q''}}{\omega(q's')\omega(q''s'')} + \frac{1}{2}\frac{\Phi^2(n_{q's'}+1)(n_{q''s''}+1)\rho\delta_{G,q-q'-q''}}{\omega(q's')\omega(q''s'')}\right], \quad (8)$$

where the factor of one half is used to avoid double counting the class II events.