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UNUSUAL SCALING OBSERVATIONS IN THE QUALITY FACTORS OF CANTILEVERED CARBON NANOTUBE RESONATORS

Ajit K. Vallabhaneni, Jeffrey F. Rhoads, Xiulin Ruan* and Jayathi Y. Murthy*
School of Mechanical Engineering
Birck Nanotechnology Center
West Lafayette, Indiana 47907
avallabh@purdue.edu

ABSTRACT

This work examines the quality factors (Q factors) of resonance associated with the axial and transverse vibrations of single-wall carbon nanotube (SWCNT) resonators through the use of molecular dynamics (MD) simulation. Specifically, the work investigates the effect of device length, diameter, and chirality, as well as temperature, on the resonant frequency and quality factor of these devices, and benchmarks the results of MD simulation against classical theories of energy dissipation. Of note are the facts that the quality factors associated with transverse vibration decrease with increasing device diameter and are largely insensitive to chirality. Additionally, quality factors increase with increasing device length for transverse vibrations, but remain almost constant for axial vibrations. The predicted size dependence of the quality factors associated with axial vibration agrees well with classical theory, if the nanoscale size effect of thermal conductivity is properly accounted for. However, the size dependence of the quality factors associated with transverse vibrations deviates significantly from classical theory.

INTRODUCTION

Since their discovery in 1991 [1], carbon nanotubes (CNTs) have become the cynosure of nanotechnology with considerable efforts being made to explore their thermal, mechanical, electrical, and optical properties. One emergent application of CNTs is in resonant nanoelectromechanical systems (NEMS) [2-4], where they can be used as enabling elements in sensors, oscillator circuits, and electromechanical signal processing systems [5-7]. The distinct utility of CNTs in these applications stems in large part from their high elastic modulus, low mass density, and high natural frequencies, which are typically in the GHz-THz range [8].

Generally speaking, the performance of a CNT resonator is constrained by the rate of energy dissipation associated with the device, which is commonly measured in terms of quality factor (Q). In most applications, a high Q is essential to optimizing performance metrics, such as device sensitivity or selectivity; and hence developing a complete understanding of dissipation in NEMS resonators is essential.

In electromechanical resonators, energy dissipation can occur through a wide variety of mechanisms [9]. Amongst these mechanisms are intrinsic processes, such as thermoelastic dissipation (TED) [10], dissipation due to electron-phonon interactions, and dissipation due to phonon-phonon interactions [11]. These mechanisms are inherent in any material and thus are omnipresent in any functional device. In contrast, there are extrinsic processes that occur due to interactions with the device's surrounding environment, such as fluidic damping and clamping losses [12]. These effects can be at least partially mitigated if proper care is taken in the course of device design and packaging.

It is important to note that a small number of prior works have considered the sources and impact of various dissipation mechanisms in CNT resonator, using experimental, analytical and numerical approaches. For example, Huttel *et al.* experimentally investigated the Q factors of resonance associated with the transverse vibration of suspended CNTs at low temperatures [13]. Likewise, a series of works have considered the temperature dependence of quality factor using molecular dynamics (MD) simulation [14, 15]. For example, in [4], Jiang *et al.* calculated the quality factors associated with the flexural vibration of CNTs and observed a $T^{-0.36}$ dependence, which deviates from classical theory. They also estimated that energy losses would in fact increase with temperature for double-walled carbon nanotubes because of interlayer interactions [14].

Despite the efforts noted above, to the best of the authors' knowledge, no prior works have investigated the impact of resonator size on energy dissipation in carbon nanotubes. This work seeks to remedy this apparent deficiency, by studying the effect of CNT size (length and diameter) on the quality factors of resonance (Q) associated with both transverse and axial vibrations through the use of MD simulations. These results are subsequently benchmarked against classical theory to highlight where further analytical efforts may be required.

METHODOLOGY

As noted in the introduction, extrinsic sources of dissipation, such as fluid dissipation, can be largely mitigated through careful device design and packaging (Note that though clamping losses can be minimized through geometric design, they are largely unavoidable), as such, this effort focuses on the dominant intrinsic sources of dissipation, namely TED and phonon-phonon effects (which are significant at high temperatures). To investigate these dissipation mechanisms, the work specifically considers a cantilevered SWCNT, which is fixed at one end and free at the other, in the absence of an adjacent substrate and surrounding medium, as shown in Figure 1.

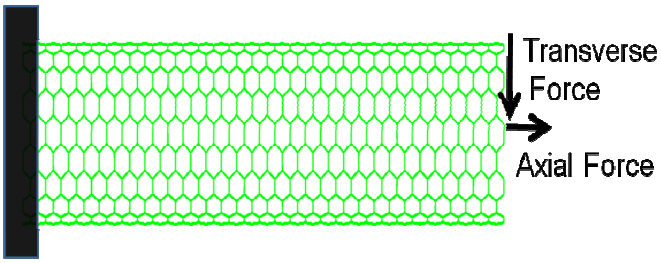

Figure 1. A REPRESENTATIVE SINGLE-WALL CARBON NANO TUBE. THE DIRECTIONS OF THE RESULTANT FORCES APPLIED DURING THE COURSE OF ANALYSIS ARE HIGHLIGHTED.

The covalent bonds between the carbon atoms which compose the system highlighted in Fig. 1 are modeled using adaptive intermolecular reactive empirical bond order (AIREBO) potentials [16], which have been shown in prior work to reproduce the elastic and thermal properties of CNT's with a high degree of accuracy [17]. Device dynamics are simulated through the use of MD simulations founded upon the LAMMPS package [18]. In each simulation, the CNT is equilibrated to a desired temperature in an NVT ensemble using a Nose Hoover thermostat for 100 ps with a 1 fs time step. The ensemble is then changed to a micro-canonical form (NVE) where the total energy of the system is kept constant. A force is subsequently applied on all of the atoms at the tip of the CNT in either the transverse or axial direction, depending on the dominant vibration mode of interest, for half the time taken for one cycle of oscillation. The force is then removed and the CNT is allowed to freely oscillate. Note that in order to ensure vibrations remain within a linear response regime; the applied forces are constrained to lead to a maximum elongation of 10% of device length.

The resonant frequency associated with the dominant mode of vibration is obtained from a Fourier transform of the time variation of the kinetic (or potential) energy. These are compared with closed-form expressions for the axial and transverse mode resonant frequencies, which are derived from elasticity theory:

$$f_{axial} = \frac{1}{4l}\sqrt{\frac{E}{\rho}}\ ;\ f_{trans} = \frac{(1.8751)^2}{2\pi}\sqrt{\frac{EI}{\rho A l^4}} \qquad (1)$$

Here, E represents Young's modulus, ρ represents mass density, l is the length of the CNT resonator, I and A are the cross-sectional moment of inertia and area of the device respectively. Note that the radius of gyration associated with the system is defined according to $K = \sqrt{I/A}$.

To compute the Q factors associated with the system of interest, variations in temperature, potential energy and displacement are considered as a function of time during the course of free vibration. In the scenario considered here, the external mechanical energy decays with time due to thermoelastic and phonon-phonon effects and is converted into internal energy, i.e., lattice energy (temperature). To quantify this effect, the variation of the displacement of the center of mass (COM) of the entire CNT in three space is used to calculate the root mean square (RMS) centroidal displacement, and subsequently Q. This procedure was previously utilized by Blencowe *et al.* [19]. Assuming linearity and a single dominant mode of vibration, the decay of the RMS displacement with time follows an exponential curve ($e^{-\zeta\omega t}$) which can be fit to calculate Q ($Q=1/2\zeta$). Here ζ is the damping ratio and ω is the angular frequency of vibration (which is equivalent to 2π multiplied by half the resonant frequency value obtained from the FFT of the kinetic energy). Q can also be calculated using external energy decay, as previously reported by Jiang *et al.* [14]. However, at high temperatures, the fluctuations in external energy are of the same order as the decay, making it hard to observe the decay clearly. By using the RMS of displacement of the center of mass, such problems do not occur, and fitting the exponential curve at high temperatures is straightforward. Accordingly, the RMS method has been adopted here, despite the fact that it offers limited value in the investigation of axisymmetric radial expansion or contraction. Note that to minimize statistical fluctuations, five independent MD simulations are performed and the results are averaged to generate each data point.

RESULTS AND DISCUSSION

In Figure 2, the dependence of frequency on length and diameter for both axial and transverse vibrations at 10 K is shown. Both of these trends follow the classical theory closely: frequency is inversely proportional to device length for axial

vibrations and to the square of device length for transverse vibrations; it is independent of diameter for axial vibrations and directly proportional to the radius of gyration for transverse vibrations.

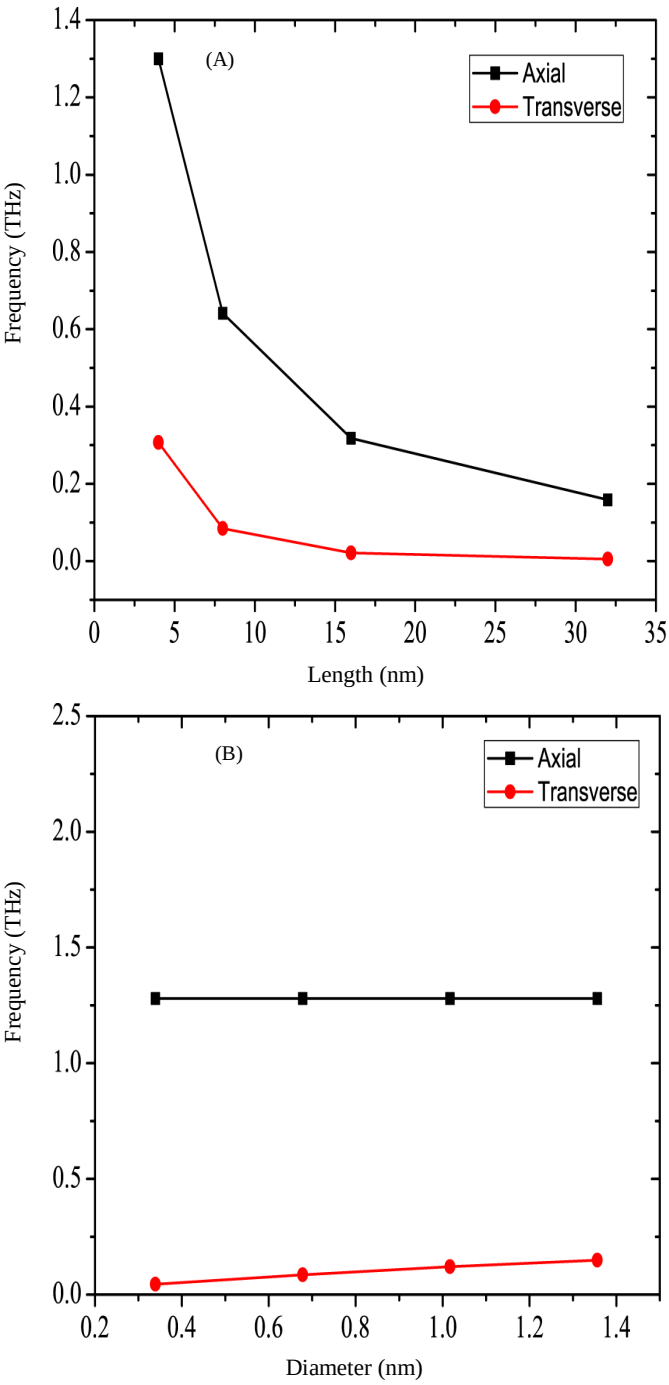

Figure 2. VARIATION OF RESONANT FREQUENCY WITH RESPECT TO SIZE. (A) RESONANT FREQUENCY AS A FUNCTION OF LENGTH, (B) RESONANT FREQUENCY AS A FUNCTION OF DIAMETER

The value of *E* calculated from the above expressions using the frequency values obtained from the fast Fourier transform (FFT) is approximately 900 GPa, which agrees with previously reported values obtained through MD simulations and molecular mechanics [20, 21]. Also of note is the fact that the arrangement of carbon atoms, armchair or zigzag, does not noticeably affect the resonant frequency. This is likely attributable to the fact that the elastic moduli of CNTs are largely independent of chirality [20]. Additionally, the frequency of both transverse and axial modes increases slightly (up to 3%) with temperature. This is in contrast to other materials for which frequency decreases with temperature. This is due to the negative coefficient of thermal expansion (CTE) in both the axial and radial directions associated with CNTs [22].

Axial-Mode Vibrations

Figure 3 shows the variation of quality factor (Q) with temperature for two distinct CNT diameters and two associated chiralities. Note that (5,5) and (9,0) CNTs have diameters which are quite close to one another, and are approximately half of those associated with (10,10) and (17,0) CNTs. As evident, Q

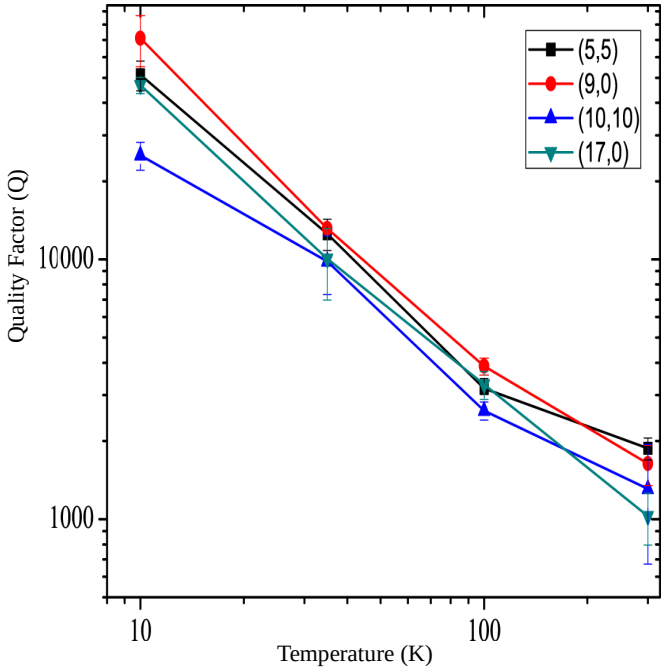

Figure 3. QUALITY FACTOR AS A FUNCTION OF CHIRALITY FOR AXIAL VIBRATIONS

decreases by approximately two orders of magnitude when temperature increases from 10 K to 300 K for each of the geometries considered. Additionally, it can be seen that the Q does not change significantly with chirality as long as the diameter is kept approximately constant. Q is also relatively independent of the orientation of atoms. Figure 4 details the Q associated with axial (longitudinal) mode CNT vibrations as a

function of length and diameter It is observed that Q is largely independent of length for (10,10) CNTs, as highlighted in Figure 4A, for a variety of different temperatures. As the length increases from 4 nm to 16 nm, the Q remains approximately constant. Potential reasons for this behavior are discussed later.

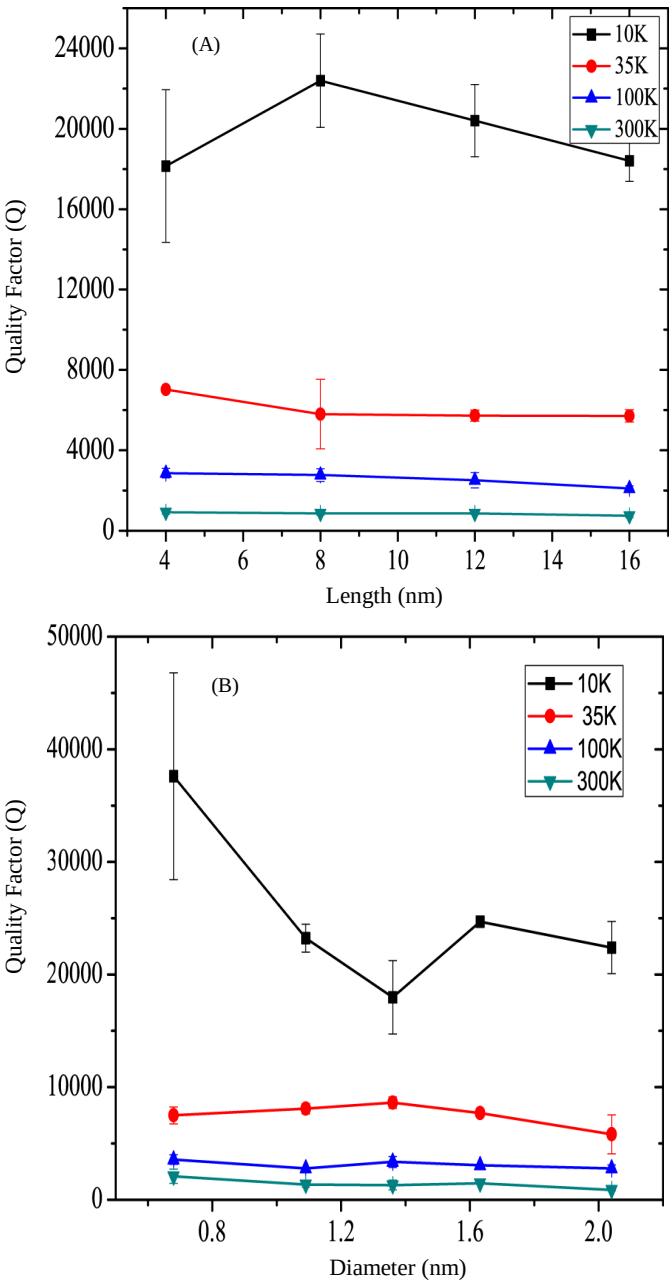

Figure 4. VARIATION OF QUALITY FACTOR FOR AXIAL VIBRATIONS WITH SIZE (A) Q AS A FUNCTION OF LENGTH (B) Q AS A FUNCTION OF DIAMETER

Figure 4B highlights Q for axial vibrations as a function of diameter at different temperatures for an 8 nm long CNT. The diameters 0.68 nm, 1.36 nm and 2.04 nm correspond to armchair CNTs of chiralities (5,5), (10,10), (15,15) respectively does not appreciably alter Q, except at low temperatures.

At 10 K, Q decreases by 40% as the diameter increases from 0.68 nm to 1.36 nm; as the diameter increases further to 2.04 nm, the Q increases again. On the whole, Q for axial mode oscillations does not vary with size significantly at least in the domain size considered here. Of the various sources of energy dissipation mentioned in the introduction, only TED is believed to be the dominant mechanism in the range of temperatures considered here. Note that Lifshitz *et al.* have proposed alternative approaches, but none of them are verified to be true [11]. TED is caused by changes in vibratory volume which results in inhomogeneous temperature changes [10]. The resulting temperature gradients induce heat flow which causes the conversion of mechanical vibration energy into thermal energy. Landau and Lifshitz [29] derived an expression for Q for longitudinal oscillations of a bar given by

$$\frac{1}{Q_{TED}} = \frac{\kappa T \alpha^2 \rho \omega}{9C^2} \tag{2}$$

where ω is the resonant frequency of the axial mode, κ is thermal conductivity, T is temperature, α is the coefficient of thermal expansion (CTE), ρ is mass density and C is heat capacity per unit volume. As per this equation, Q should be independent of diameter and chirality for axial vibrations because the parameters κ, α, C vary only slightly with diameter. Furthermore, as shown in Figure 2, ω is independent of diameter. If one considers length dependence, ω varies as $1/L$ and all other parameters in Equation (2) are constant with length except for thermal conductivity (κ). Since, the domain lengths considered here are much less than the mean free path, κ increases with length almost linearly from 4 nm to 16 nm because of the finite size effect [17, 23]. This implies that the product of κ and ω will remain almost constant. Thus, Q should be approximately constant with length, which matches our observations in Figure 4. However, a slight deviation from this behavior is observed at 10 K and the reasons for this are not yet clear. Once the diffusive regime is reached, (i.e., for lengths greater than the mean free path) κ no longer increases with length; thus one would expect the quality factor to decrease with increasing length in this limit. To the best of the authors' knowledge, no experiments have been done on nanoscale axial mode vibrations to verify this behavior. In summary, the Q for axial mode vibrations is independent of size (at least within the parameter ranges considered herein) and increases with decreasing temperature.

Transverse-Mode Vibrations

The procedure described above is repeated for transverse mode vibrations, wherein the force is applied perpendicular to the axis of CNT. As highlighted by Figure 5, the Q for transverse mode vibrations is found to be independent of

chirality. Only the diameter of the CNT affects the quality factor of transverse mode vibration but not its chirality. Figure 6 shows the variation of Q with increasing diameter for a variety of temperatures. It is clear that Q in this case decreases with increasing diameter for all of the temperatures under consideration. The decrease of Q with diameter at high temperatures is demonstrated in Figure 6B. The dependence of Q on diameter (b) follows the power law:

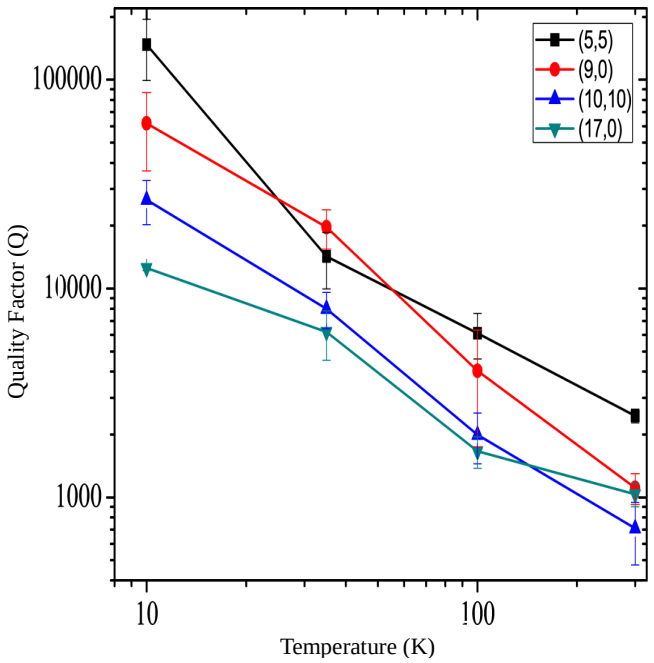

Figure 5. VARIATION OF QUALITY FACTOR WITH CHIRALITY FOR TRANSVERSE VIBRATIONS

But the dependence of size (length & diameter) is stronger in the case of transverse mode vibrations unlike the axial. $Q \sim b^{-\mu}$ where μ varies between 1.4 and 1.6. In other words, the damping increases with frequency. Figure 7 shows the variation of Q for the transverse mode of vibration as a function of length for a (10,10) CNT. It can be seen that Q increases with length in this case. As can be seen in this figure, for various temperatures, Q increases with length as per the law: $Q \sim L^{\theta}$ where θ varies from 1 to 1.5. Both of these trends are in contrast with the trends observed for the case of axial vibrations. TED can play a significant role in this case as well since temperature gradients are developed across the diameter due to alternating tensile and compressive forces. This results in an irreversible loss of mechanical energy.

Zener studied TED (dubbed internal friction) in flexural vibrations in 1938, and in a series of papers [24-27] derived an expression for Q_{TED} for a rectangular beam given by:

$$\frac{1}{Q_{TED}} = \frac{E\alpha^2 T_0}{C_p} \frac{\omega\tau_z}{1+(\omega\tau_z)^2} \qquad (3)$$

Where E is Young's modulus, α is coefficient of thermal expansion, C_p is specific heat, T_0 is the temperature.

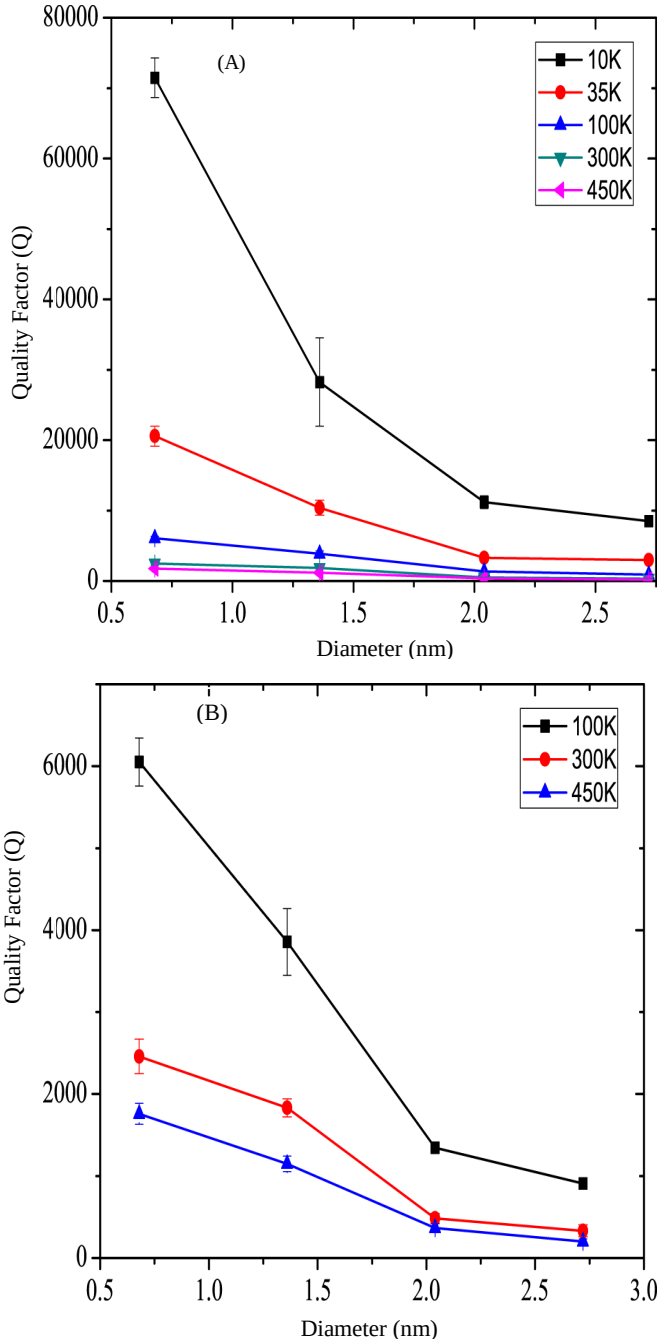

Figure 6. VARIATION OF QUALITY FACTOR WITH DIAMETER FOR TRANSVERSE VIBRATIONS (A) AT ALL TEMPERATURES UNDER CONSIDERATION (B) AT HIGH TEMPERATURES (ZOOMED VIEW)

The relaxation time (τ_z) is given by

$$\tau_z = \frac{b^2}{\pi^2 \chi}$$

Here b is the width (approximated as diameter here) of the beam and χ (= κ/ρC) is the thermal diffusivity of the solid.

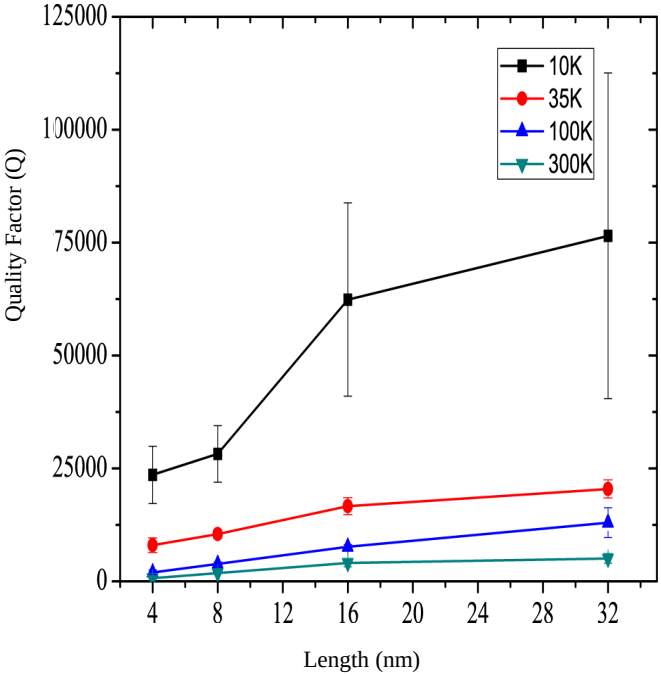

Figure 7. VARIATION OF QUALITY FACTOR WITH LENGTH FOR TRANSVERSE VIBRATIONS

The values of relaxation time and its product with frequency, for transversely vibrating CNTs of different chiralities and 4 nm length are listed in Table 1.

Table 1. VALUES OF FREQUENCY AND RELAXATION TIMES FOR DIFFERENT CHIRALITIES

Chirality	ω (THz)	τ_z (fs)	$\omega\tau_z$ (x10^{-3})
(5,5)	0.181	3.258	3.706
(10,10)	0.308	13.032	25.22
(15,15)	0.394	29.324	72.594

In the domain size considered, the product $\omega\tau_z$ is less than 1, which means that relaxation time is shorter than the period of oscillation, implying that the oscillations are isothermal. Increasing the length decreases the product further where as increasing the diameter could get the value closer to unity. With this approximation, the expression for Q in Equation (3) reduces to

$$\frac{1}{Q_{TED}} = \frac{E\alpha^2 T}{\rho C}\omega\tau = \frac{E\alpha^2 T}{\rho C}\frac{\omega b^2}{\pi^2 \chi}$$
$$= \frac{E\alpha^2 T}{\rho C}\frac{\omega b^2}{\pi^2 (\kappa/\rho C)} = E\alpha^2 T \frac{\omega b^2}{\pi^2 \kappa} \quad (4)$$

Based on Equation (3), Photiadis *et al.* predicted that Q will increase with temperature because of the change in sign of CTE [28], but this trend was not reflected in their experiments. Even though CNTs also exhibit a change of sign in CTE as temperature increases [22], such anomalous behavior of Q with temperature is also not observed in simulation. As per Equation (4), Q should decrease rapidly with increasing diameter since Q ~ $1/b^2$. This is especially true given that the resonant frequency (which is directly proportional to diameter) is also inversely proportional to Q. From the results presented here, Q is observed to have a weaker dependence on diameter (~ $1/b^{1.5}$) than expected. Lifshitz *et al.* proposed that the relaxation time varies linearly with diameter (τ ~ b/ν, where ν is the phonon group velocity) rather than quadratically [10], within the ballistic range, which might help explain this deviation; but there is no comprehensive justification in this regard. Similar deviations are seen in the dependence of Q on length. Here, frequency is the only term which varies with length; therefore one would expect Q to increase quadratically with that parameter (~ L^2). However, the observed dependence is only linear (~ L). This could be a byproduct of multimode contributions or increased coupling between axial and transverse modes of vibration. A more rigorous spectral analysis has to be done to gain more insights in this direction. Finally it should be noted that the authors have also looked at phonon-phonon dissipation and the inclusion of that effect doesn't seem to offer a better correlation between theory and simulation.

In summary, to obtain higher Q for transverse mode vibrations, devices of high aspect ratio (L/D); i.e., larger length and smaller diameter, should be utilized at low temperatures.

CONCLUSIONS

In summary, this work has considered the Qs associated with axial and transverse mode vibrations in CNTs of different lengths and diameters using MD. Though there has been speculation in the literature that Q may increase with temperature, such behavior has not been observed. Simulation results indicate that resonant frequencies scale with size, in the same manner as predicted by classical theory. The values of Q obtained for axial oscillations were largely independent of size and agreed well with Landau's classical theory. The authors speculate that Q changes with length in the diffusive regime, but this must be verified through future simulations. In the case of transverse mode oscillations, Q was shown to be quite sensitive to the dimensions of the system. Variations with size were observed to be in qualitative, but not quantitative, agreement with Zener's classical theory of TED. The authors conclude that

none of the existing classical theories are successful in fully explaining these unusual scaling laws.

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