Observation of nonclassical scaling laws in the quality factors of cantilevered carbon nanotube resonators

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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 simulations against classical theories of energy dissipation. The quality factor (Q) associated with transverse vibration is found to increase with increasing device length ($Q \sim L^{\theta}$, where $0.8 < \theta < 1.4$) and decrease with increasing device diameter ($Q \sim D^{-\mu}$, where $1.4 < \mu < 1.6$), while the Q associated with axial vibration is almost independent of length and diameter. We show that to accurately predict temperature dependence of Q, the external and internal energies need to be properly decomposed, and temperature quantum correction should be performed. For both vibrational modes, Q shows a temperature dependence $Q \sim T^{-\alpha}$, where $\alpha > 1$ when below Debye temperature due to quantum effects, and Q gradually recovers the classical T^{-1} dependence when above Debye temperature. Our temperature dependence is in contrast to prior studies that suggested $Q \sim T^{-\beta}$, where $0 < \beta < 1$. The observed size and temperature dependencies by us have many deviations from existing classical theories of energy dissipation, possibly due to phonon confinement effects in these nanostructures and temperature quantum effects. © *2011 American Institute of Physics*. [doi:10.1063/1.3611396]

I. INTRODUCTION

Since their discovery in 1991,[1] carbon nanotubes (CNTs) have become a significant focus of research in nanotechnology with considerable efforts being made to explore their thermal, mechanical, electrical, and optical properties. One emerging 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 to 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 therefore 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] Among these mechanisms are intrinsic processes such as thermoelastic dissipation (TED),[10] dissipation due to electron-phonon interactions,[11] and dissipation due to phonon-phonon interactions.[12] 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.[13] These effects can be at least partially mitigated if proper care is taken in the course of device design and packaging.

A small number of prior works has considered the sources and impact of various dissipation mechanisms in CNT resonators, using experimental, analytical, and numerical approaches. For example, Huttel *et al*. experimentally investigated the Q of resonance associated with the transverse vibration of suspended CNTs at low temperatures.[14] Likewise, a series of papers has considered the temperature dependence of quality factor using molecular dynamics (MD) simulations.[15–17] For example, 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.[15] A recent paper by Greaney *et al*. demonstrates the possibility of anomalous dissipation in SWCNTs because of the interaction between various mechanical modes.[18]

Despite the efforts noted in the preceding text, to the best of the authors' knowledge, no prior works have investigated the impact of CNT size, including length and diameter, on energy dissipation. This topic is of distinct interest because it is not clear whether the classical scaling laws on resonator size, developed using macroscopic continuum theories, are still valid in nanoscale systems such as the CNT resonator considered here. In an attempt to address this, this work studies the effect of CNT size (length and diameter) on

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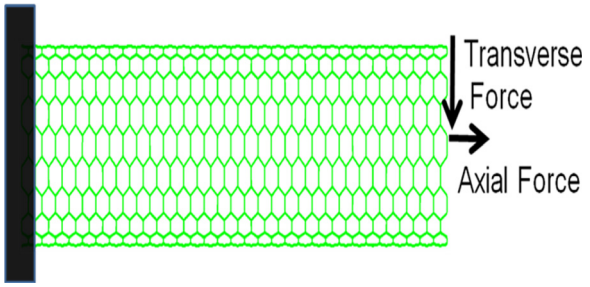

FIG. 1. (Color online) A representative single wall carbon nanotube. The directions of the resultant forces applied during the course of analysis are highlighted.

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 non-classical behavior may occur.

As noted earlier, extrinsic sources of dissipation, such as fluid dissipation, can be largely mitigated through careful device design and packaging (note that although clamping losses can be minimized through geometric design, they are largely unavoidable). Therefore 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 Fig. 1.

II. METHODOLOGY

The covalent bonds between the carbon atoms that compose the system highlighted in Fig. 1 are modeled using adaptive intermolecular reactive empirical bond order (AIREBO) potentials.[19] Device dynamics are simulated through MD simulations using the LAMMPS package.[20] In each simulation, the CNT is equilibrated to a desired temperature in an NVT ensemble using a Nose-Hoover thermostat for 100 ps with 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 by applying the force on the free end, the vast majority of energy goes in to the principal transverse (or axial) mode of vibration as verified in the supplemental information.[33] Also note that to ensure that 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. To compute Q, the variations in external energy and displacement need to be considered as a function of time during the course of free vibration. In the scenario considered here, the external (mechanical) energy decays with time either due to thermoelastic and/or phonon-phonon effects and is converted into internal (thermal) energy of the system.

Q has been most commonly calculated using the decay rate of the external energy, as previously practiced in the literature.[15] However, MD simulations can only directly output the total energy that is a summation of external (mechanical) energy and internal (thermal) energy, and the decomposition of the external and internal energies is not trivial. In the above-mentioned studies,[15–17] the external and internal energies are not clearly separated; this seems to lead to a slower decay of "external energy" than what actually takes place. This results in the quality factors being generally overestimated. The overestimation is worse at higher temperatures due to the correspondingly larger internal energy. The internal energy must be subtracted to estimate the Q of the corresponding transverse (or axial) mode accurately. Also, at high temperatures, the fluctuations in external energy are of the same order as the decay, making it hard to observe the decay clearly.

Alternatively, the Q factor can be calculated using the variation of the displacement of the center of mass (COM) of the entire CNT in three dimensional space, as previously utilized by Blencowe *et al.*[21] Assuming linearity and a single dominant mode of vibration, the decay of the root mean square (RMS) centroidal 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). By tracking the motion of the COM, the bulk motion corresponding to the principal transverse (or axial) mode is isolated while the thermal motion of atoms is averaged out. This eliminates the inconvenience of decomposing internal and external energies that is essential in the energy decay approach. Also, the exponential decay of the RMS displacement 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.

III. RESULTS

The frequencies obtained from Fourier transform are compared with closed-form expressions for the axial and transverse mode resonant frequencies, which are derived from elasticity theory:

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

Here, E represents Young's modulus, ρ represents mass density, l is the length of the CNT resonator, and 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 $K = \sqrt{I/A}$. In Fig. 2, the dependence of frequency on length and diameter for both axial and

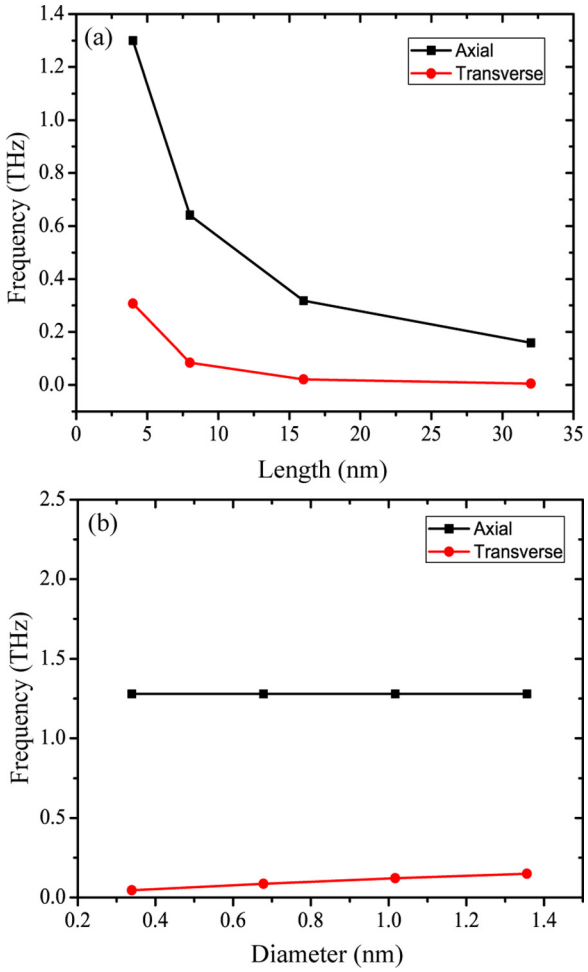

FIG. 2. (Color online) Variation of resonant frequency with respect to size at 10 K. (a) resonant frequency as a function of length for a (10,10) CNT and (b) resonant frequency as a function of diameter.

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. The value of E calculated from the preceding expressions using the frequency values obtained from the fast Fourier transform (FFT) is approximately 900 Gpa; this agrees with previously reported values obtained through MD simulations and molecular mechanics.[22,23] 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.[22] In addition, 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 possibly due to the negative coefficient of thermal expansion (CTE) in both the axial and radial directions associated with CNTs.[24]

The typical decay of the RMS of the displacement of the COM of a 4 nm long (10,10) CNT at 100 K is plotted in Fig. 3. This exponential decay rate is used to calculate the Q as mentioned earlier. Figure 4(a) shows the variation of

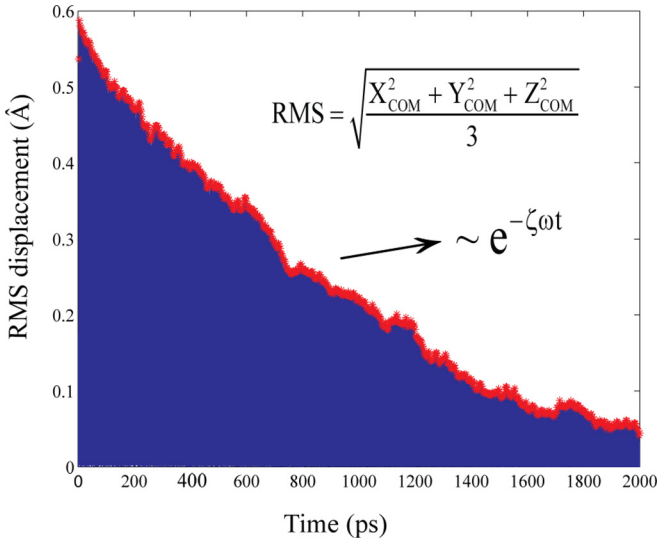

FIG. 3. (Color online) Typical decay of RMS of displacement of the center of mass (COM) with time in a 4 nm long (10,10) CNT at 100 K.

quality factor (Q) with the temperature in the MD simulations for two distinct CNT diameters and two associated chiralities. Note that (5,5) and (9,0) CNTs have diameters that are quite close to one another and are approximately half of those associated with (10,10) and (17,0) CNTs. It is evident that as the temperature increases, Q decreases as T^{-1} in accordance with classical theory for each of the geometries considered. The exponent on T, i.e., −1 is different from the value of −0.36 reported by Jiang *et al.*,[15] who estimated Q based on the decay of external energy. As noted earlier, this approach can be misleading as the "external energy" utilized there actually includes contributions from the other mechanical and thermal modes, the initial thermal energy in addition to the principal mechanical energy. Accordingly, the Q factor thus was overestimated, especially at higher temperatures, which causes the weak temperature dependence. To validate this conjecture, we used a low-pass filter (based on the known frequency of the mechanical mode) to isolate the external energy decay corresponding to the dominant mechanical mode. The magnitude of Q obtained using this approach is virtually identical to that of Q calculated using the RMS method (Please see the supplemental information[33] for more details in this regard).

However, because the temperatures in our MD simulations (T_{MD}) are lower than the Debye temperature of CNTs (475 K),[24] quantum correction needs to be performed by equating the total energy of the system from the classical and quantum theories such as done by Lukes *et al.*[25] Detailed procedure of quantum correction is presented in the supplemental information. The corrected temperatures corresponding to classical temperatures in our MD simulations (T_{MD}) 10 K, 35 K, 100 K, 300 K are 82 K, 128 K, 201 K, and 376 K, respectively. The variation of Q with temperature after quantum correction is plotted in Fig. 4(b). In general, Q varies inversely with phonon population indicating phonon–phonon coupling is probably responsible for energy dissipation. Below Debye temperature, the temperature dependence on Q is stronger than the T^{-1} relation due to quantum effects, and as T_{MD} reaches

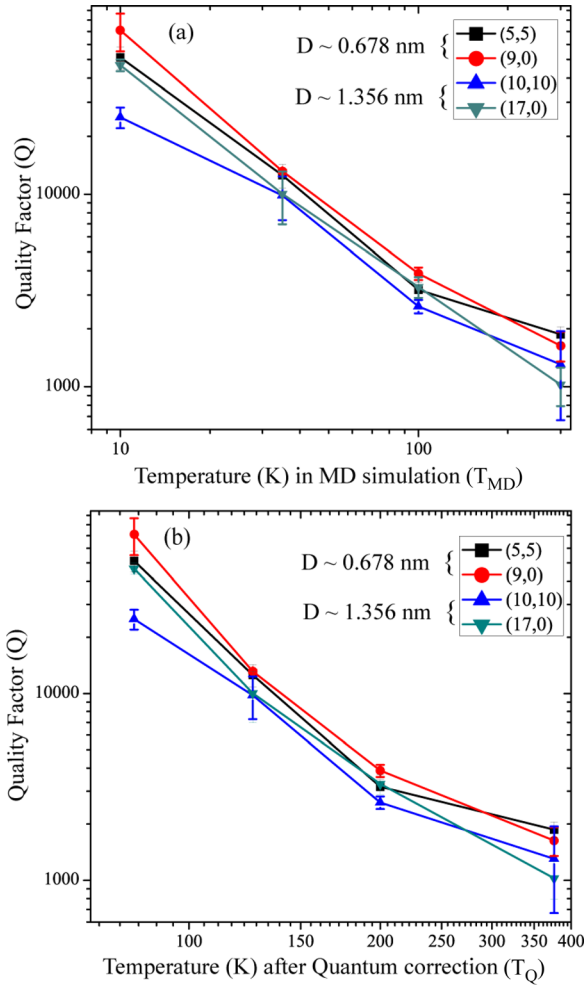

FIG. 4. (Color online) Quality factors plotted as a function of temperature for axially vibrating CNTs in (a) classical regime and (b) quantum regime (after quantum correction).

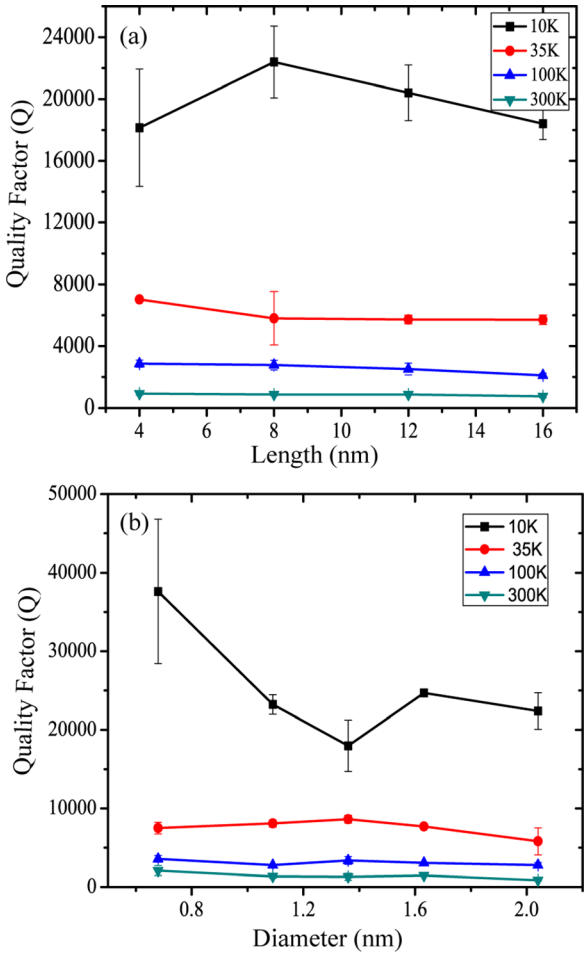

FIG. 5. (Color online) Variation of quality factor for axial vibrations with size (a) Q plotted as a function of length and (b) Q plotted as a function of diameter.

the Debye temperature, it should gradually recover the classical T^{-1} trend. More details are included in the supplemental information[32] in this regard. Temperature quantum correction was omitted in prior MD simulations of Q of carbon nanotubes or graphene,[15–17] which is another source of the inaccurate temperature dependence. A theoretical study by Johannes *et al*. also showed a similar temperature dependence.[33]

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 5 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 Fig. 5(a), for a variety of different temperatures. As the length increases from 4 nm to 16 nm, the Q remains approximately constant. Note that to minimize statistical fluctuations, five independent MD simulations are performed, and the results are averaged to generate each data point.

Figure 5(b) presents Q for axial vibrations as a function of diameter at different temperatures for an 8 nm long CNT. The diameters 0.68 nm, 1.08 nm, 1.36 nm, 1.63 nm, and 2.04 nm correspond to armchair CNTs of chiralities (5,5), (8,8), (10,10), (12,12), and (15,15), respectively. It can be observed that the diameter of the CNT 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 significantly with size at least for the domain sizes considered here. To the best of the authors' knowledge, no experiments have been done on nanoscale axial mode vibrations yet to demonstrate this behavior.

The procedure described in the preceding text is repeated for transverse mode vibrations, wherein the force is applied perpendicular to the axis of the CNT. As can be seen in Fig. 6, the diameter of the CNT affects the quality factor of transverse mode vibration significantly but not its chirality. The temperature dependence is very similar to that of the axial mode. Figure 7(a) shows the variation of Q for the transverse mode of vibration as a function of length for a (10, 10) CNT. As can be seen, for various temperatures, Q increases with length approximately following the law $Q \sim L^{\theta}$ where θ varies from 0.8 to 1.4. Figure 7(b) demonstrates the variation of Q with diameter for temperatures from 10 K to 300 K. It is clear that Q in this case decreases with increasing diameter for all of the temperatures under consideration. The dependence

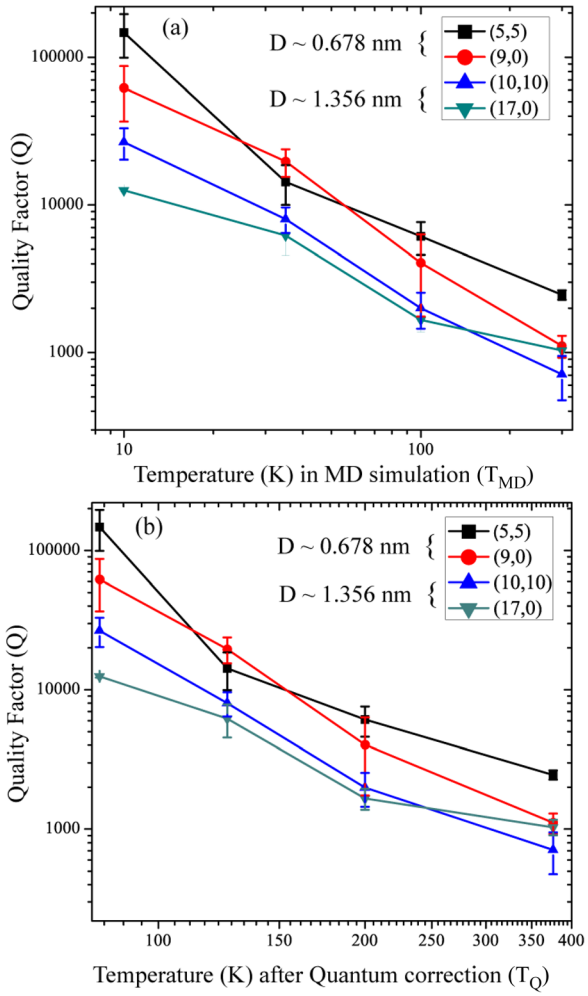

FIG. 6. (Color online) Quality factors plotted as function of temperature for transverse vibrations in (a) classical regime and (b) quantum regime (after quantum correction).

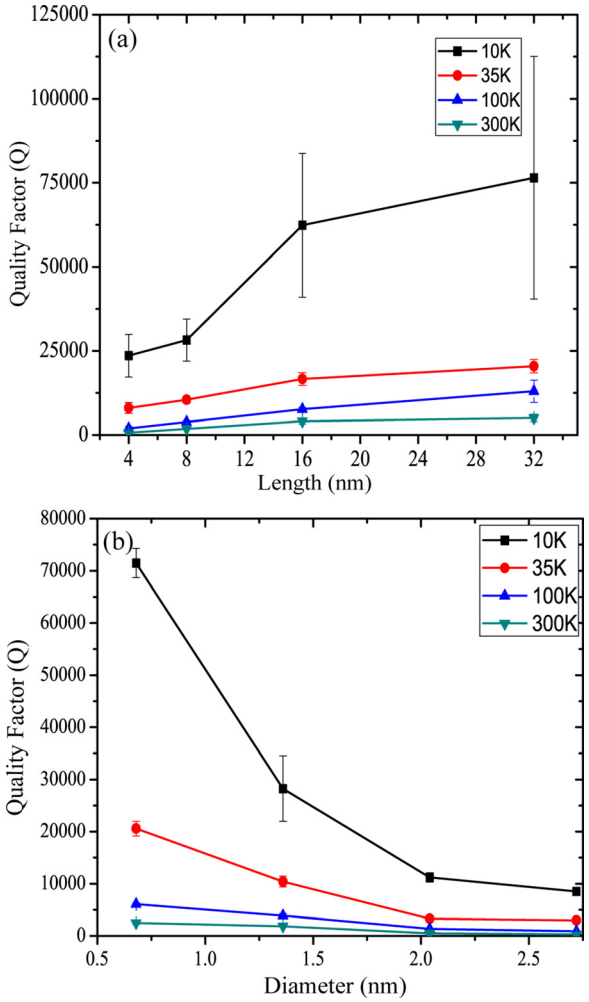

FIG. 7. (Color online) Variation of quality factor for transverse vibrations with size (a) Q plotted as a function of length and (b) Q plotted as a function of diameter

of Q on diameter (D) follows the power law: $Q \sim D^{-\mu}$ where μ varies between 1.4 and 1.6. It is evident that the dependence of Q on size (length and diameter) is stronger in the case of transverse mode vibrations than the axial mode.

These results indicate that to obtain higher Q for transverse mode vibrations, devices with high aspect ratio (L/D); i.e., larger length and smaller diameter, should be used at temperatures as low as possible.

IV. COMPARISON WITH CLASSICAL THEORY OF ENERGY DISSIPATION

The following section includes a brief summary of classical theories that are relevant to the present discussion and comparison of previous results obtained from MD simulations against these theories. Of the various sources of energy dissipation mentioned in the introduction, TED and phonon–phonon damping are believed to be the dominant mechanisms in the range of temperatures considered here. TED is caused by changes in vibratory volume; this results in inhomogeneous temperature changes.[10] The resulting temperature gradients induce heat flow, which causes the conversion of mechanical vibration energy into thermal energy. The expression for quality factor in this case is given by

$$\frac{1}{Q} = \frac{E\alpha^2 T_0}{C_p} \frac{\omega\tau}{1+(\omega\tau)^2}, \tag{2}$$

where E is the Young's modulus, α is the coefficient of thermal expansion, C_p is the specific heat, T_0 is the temperature, and τ is the relaxation time given by

$$\tau = \frac{r^2}{\pi^2 \chi}. \tag{3}$$

Here, r is the length scale over which temperature gradients are developed, and χ $(= \kappa/\rho C)$ is the thermal diffusivity of the material. For axial vibrations, r is equal to half the wavelength (λ) of the wave, and for transverse vibrations, it is roughly equal to thickness (diameter) of the tube. Based on Eq. (2), Photiadis *et al.* predicted that Q would increase with temperature because of the change in sign of CTE,[26] but this trend was not reflected in their experiments. Even though CNTs also exhibit a change of sign in CTE as temperature increases, this anomalous behavior of Q with temperature is also not observed in our simulations.

Substituting the preceding expression for τ in Eq. (2) and using the relation $\lambda = c/f$ where c is the velocity of sound and f is the frequency of excited mechanical mode, the expression in Eq. (2) for axial vibrations in the adiabatic limit ($\omega\tau \gg 1$) simplifies to the following:

$$\frac{1}{Q_{TED}} = \frac{\kappa T \alpha^2 \rho \omega}{9C^2}, \tag{4}$$

where ω ($= 2\pi$f) is the resonant frequency, κ is the 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.[11] 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 and ω is also diameter independent as shown in Fig. 2. As length increases, frequency decreases ($\sim 1/L$) and Q is expected to increase almost linearly as a result of this equation. However, the simulation data show that Q is almost independent of L. This may be due to the fact that the thermal transport in the system is in the ballistic regime. The length of CNTs considered in our simulations is much less than the phonon mean free path, and as a result κ increases with length almost linearly from 4 nm to 16 nm. 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 Fig. 5. By taking into account this non-classical correction into Eq. (4), our simulation results are in good agreement with Eq. (4).

The expression for quality factor for transverse vibrations was derived by Zener for rectangular reeds[27–29] based on the assumptions that heat flow is one dimensional, and only one thermal mode is coupled to the mechanical mode. For the domain size considered here, the product $\omega\tau \ll 1$; corresponding to the isothermal limit. In this limit, the expression for Q simplifies to the following:

$$\frac{1}{Q_{TED}} = \frac{E\alpha^2 T_0 \omega D^2}{\pi^2 \kappa}, \tag{5}$$

where D is the diameter of the nanotube. As per this relation, Q should decrease rapidly with increasing diameter because $Q \sim 1/D^2$. 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 ($\sim 1/D^{1.5}$ approximately) than predicted by this theory. Similar deviations are seen in the dependence of Q on length. Here, frequency is the only term that varies with length; therefore one would expect Q to increase quadratically with that parameter ($\sim L^2$). However, the observed dependence is almost linear ($\sim L$). Hence, the thermoelastic dissipation theory can only partially explain the presented results.

Thermoelastic dissipation mechanism is considered relevant mostly when the wavelength of the acoustic mode is much greater than the phonon mean free path. Lifshitz *et al.* have proposed alternate approaches for the ballistic regime, but there is no comprehensive justification given in this regard.[10] In addition to the above-mentioned mechanism, the interaction between acoustic waves with thermal phonons gives rise to another intrinsic mechanism known as phonon–phonon dissipation. Q in this case is given by the expression:

$$\frac{1}{Q_{ph-ph}} = \frac{CT\gamma^2}{\rho \upsilon^2} \frac{\omega\tau_{ph}}{1 + \left(\omega\tau_{ph}\right)^2}, \tag{6}$$

where γ is the Gruneisen parameter, C is the specific heat per unit volume, ρ is the density, υ is the sound velocity, and the phonon relaxation time τ_{ph} is given by

$$\kappa = \frac{1}{3} C \nu_D^2 \tau_{ph}, \tag{7}$$

where κ is thermal conductivity and ν_{D} is the Debye sound velocity. This expression is valid only in the diffusive range ($\omega\tau_{\mathrm{ph}} \ll 1$) where the phonon mean free path is less than the wavelength. This was derived by Akheiser based on the assumption that the acoustic wave interacts with the whole ensemble of thermal phonons.[30]

Here, the phonon mean free path is greater than the wavelength of the acoustic mode ($\omega\tau_{\mathrm{ph}} \gg 1$). Phonon-phonon interaction in this range is referred to as the Landau–Rumer effect[31] and Q is given by:

$$\frac{1}{Q} = \frac{\pi^4 \gamma^2 k^4 T^4}{30 \rho v^5 h^3}, \tag{8}$$

where γ is the Gruneisen parameter, k is the Boltzmann constant, ρ is the density, ν is the wave velocity, and h is Planck's constant. Here the acoustic wave can be considered as interacting with individual thermal phonons. This expression has been derived by assuming three phonon interactions (one acoustic and two thermal). The fourth order temperature dependence is a result of quantum effects at extremely low temperature, which can agree with our MD simulation after temperature quantum correction. However, this scaling law indicates no size dependence at all; this is in contrast to the aforementioned simulation results. Further analysis must be performed to explain these nonclassical behaviors in this regard.

V. CONCLUSIONS

In summary, this work has considered the quality factors associated with axial and transverse mode vibrations in CNTs of different lengths and diameters using classical molecular dynamics simulations. The included 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 are largely independent of size and this trend agrees well with Landau's classical theory for TED (if the nanoscale size effect on κ is included) and also the Landau–Rumer effect. The authors speculate that Q varies with length in the diffusive regime, but this must be verified through future simulations. In the case of transverse mode oscillations, Q is shown to be quite sensitive to the dimensions of the system. Variations with size are observed to be in qualitative, but not quantitative, agreement with Zener's classical theory of TED. For both vibrational modes, Q shows a temperature dependence $Q \sim T^{-\alpha}$, where

$\alpha > 1$ when below Debye temperature due to quantum effects, and Q gradually recovers the classical T^{-1} dependence when above Debye temperature. Our temperature dependence is in contrast to prior studies where the external and internal energy were not separated, and temperature quantum correction was not performed. The authors conclude that none of the existing classical theories can fully explain these unusual scaling laws and further development of theories at the nanoscale is warranted.

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