A band-pass filter approach within molecular dynamics for the prediction of intrinsic quality factors of nanoresonators

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(Received 7 March 2012; accepted 24 August 2012; published online 1 October 2012)

The temperature and frequency dependence of the flexural mode quality factors (Q) of doubly clamped single wall carbon nanotube resonators are calculated using classical molecular dynamics simulations. The validity of the various methods available in the literature for calculating Q based on the temporal response of the system during ring-down is discussed and the discrepancies associated with the methods are explained. A new approach based on band-pass filtering is proposed for calculating Q, which reveals classical temperature dependence ($Q \sim T^{-1}$) in contrast to the previously reported results ($Q \sim T^{-\beta}$, $0 < \beta < 1$). It is shown that the Q estimated from the temporal response is in good agreement with the Q estimated from frequency response. This work also demonstrates that the proposed method is particularly advantageous when multiple modes are simultaneously excited within the linear regime. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4754450]

I. INTRODUCTION

Since their discovery in 1990,[1] carbon nanotubes (CNTs) have garnered significant interest because of their unique thermal,[2] mechanical, and optical properties.[3] These excellent properties coupled with recent experimental advancements at the micro and nanoscales have motivated researchers to use them for a variety of novel applications.[4–6] One such application is nanoelectromechanical systems (NEMS)[7] where CNTs are one of the most sought after materials due to their low mass density and high Young's modulus, which result in very high natural frequencies (GHz-THz).[8–10] In addition to frequency, the performance of these resonant devices is commonly measured in terms of the quality factor (Q), which is defined as the ratio of energy stored to energy lost per cycle of motion. A higher Q is generally desirable as it results in higher efficiency and greater device sensitivity.

While the miniaturization of scale can lead to high Q, experimental measurements on CNT resonators at room temperature have yielded surprisingly low Q (<100). The highest Q ever reported for the fundamental flexural mode of a single wall CNT (SWCNT) resonator below room temperature was 140 670 at 20 mK.[9] The intrinsic losses which are caused by the inherent thermal and mechanical properties of the device set the upper limit for Q in any resonator. While extrinsic losses like air damping and clamping losses are commonly present, they can be controlled to a certain degree by taking proper care in design and fabrication. Given the dominance of extrinsic losses in experiments, molecular dynamics (MD) simulations offers an opportunity to study the intrinsic dissipation with no external losses, and to establish the upper limits on the maximum achievable Q. MD simulations have been used previously to study the intrinsic dissipation mechanisms in nanotubes [11,12] and nanowires.[13] In these works, Q has been calculated using the decay rate of the external energy ($E_n = E_{ext}(1–2\pi/Q)^n$), where E_n is the external potential (or kinetic) energy and n is the number of cycles. This resulted in a non-classical temperature dependence ($Q \sim T^{-\beta}$ where $0 < \beta < 1$).

In the current work, we show that the reported Qs in the existing literature are overestimated because of an incorrect definition of external energy. We introduce a new band-pass filter (BPF) approach that can correctly render the external energy by removing the thermal noise in the temporal response of energy. Using the correct definition of the external energy (and also the displacement of the center of mass), we show that the temperature dependence of Q is indeed classical. We then compare the BPF approach to the center of mass (COM) approach and find that the BPF approach is capable of predicting the quality factors of all modes under multi-mode excitations within the linear regime. The BPF approach is then used to calculate the flexural-mode quality factors in doubly clamped carbon nanotube resonators, in order to study the frequency dependence. The motivation behind the present work is to compare the different methods that can be used in estimating Q and to study the Q's dependence on temperature and frequency (including higher-order modes). In view of the emerging interest in studying the intrinsic damping characteristics of nanowires and nanotubes using MD simulations, we believe that the approach introduced here will serve as an effective tool to accurately calculate Q in the future works. Note that the authors have previously investigated the non-classical size dependence of Q in cantilevered SWCNT resonators in Ref. 14.

II. SIMULATION METHODOLOGY

A pristine 16 nm long CNT with fixed boundary conditions is considered here [see Fig. 1(a)]. Since the focus of the present study is the intrinsic damping characteristics of the CNT, no substrate or surrounding medium is included.

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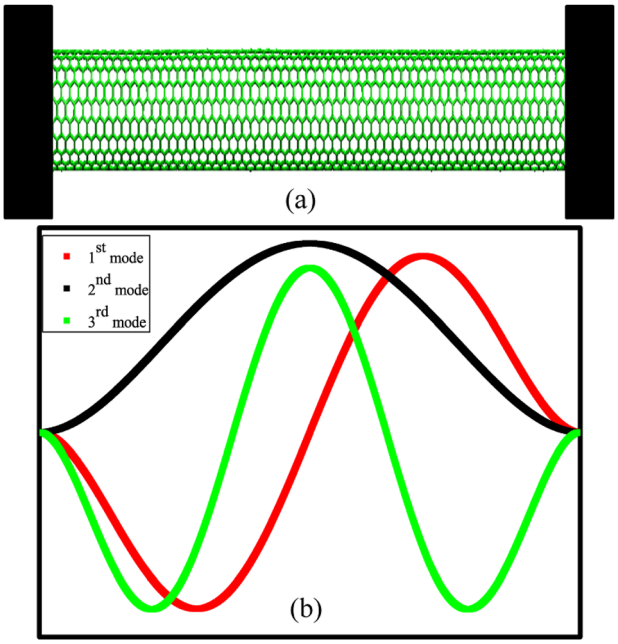

FIG. 1. (a) A (10,10) 16 nm long SWCNT with fixed-fixed boundary conditions. (b) Mode shapes of the first three transverse modes.

Figure 1(b) shows the mode shapes of the first, second, and third flexural modes of a typical fixed-fixed beam. In our simulations, the interactions between carbon atoms are modeled using the adaptive intermolecular reactive empirical bond order (AIREBO)[15] potential and all the simulations are carried out using the LAMMPS package.[16] Initially, free boundary conditions are applied, and the CNT is equilibrated to a desired temperature using a Nose-Hoover thermostat in an NVT ensemble [where the number of atoms (N), volume (V), and temperature (T) are constant] for 200 ps with a time step of 1 fs. Then atoms at both ends of the CNT are fixed and the system is equilibrated again for 200 ps. The ensemble is then switched to a micro-canonical form (NVE) where the total energy of the system is kept constant. Subsequently, a force is applied at the atoms in the center of the CNT for half of an oscillation period. The force is then removed and the temporal response of the system's kinetic energy and the displacement of the COM are monitored to estimate Q.

Carrying out the ring-down in a constant energy (NVE) ensemble has the advantage that the energy lost from the mechanical motion is converted into and stored as the internal energy of the system. To ensure that the vibrations remain within a linear response regime, the applied forces are constrained to cause a maximum displacement of 4% of the length. To excite the higher order flexural modes (2nd and 3rd), forces are applied at the antinodes of the mode shape as will be shown below in Fig. 5(a). The resonant frequency of the dominant mode of vibration is obtained from the Fourier transform of the time response of the kinetic (or potential) energy.

III. EXTERNAL ENERGY AND THE TEMPERATURE DEPENDENCE OF Q

As mentioned previously, here Q is calculated based on the temporal response of the CNT. The external energy and the root mean square (RMS) of the COM have been used to calculate the value of Q during the ring-down of the system.[17] Note that the external energy of the system by definition is the energy corresponding to the excited mechanical mode alone and it should decay to zero once the system reaches equilibrium. However, the MD simulation does not output this quantity directly. Rather, it provides the total kinetic energy of the system, i.e.,

$$E_k(t) = E_{ek}(t) + E_{ik}(t) = E_{ek}(t) + [E_{ik,sm}(t) + E_{ik,n}(t)]. \quad (1)$$

Here, $E_k(t)$ is the total kinetic energy after the excitation of a mode as a function of time at any particular temperature of interest. This energy is the summation of the external kinetic energy for the flexural mode, denoted as $E_{ek}(t)$, and the internal kinetic energy due to thermal vibrations, denoted as $E_{ik}(t)$. $E_{ek}(t)$ oscillates at a frequency of $2f_n$, where f_n is the natural frequency of the flexural mode and is usually in the GHz range. It reaches the minimum value (zero) at the maximum displacement and the maximum value when passing the equilibrium position. The amplitude of the oscillation decays with time, eventually approaching zero after all of the mechanical energy is lost to thermal energy. In contrast, $E_{ik}(t)$ oscillates at much higher frequencies that correspond to phonon frequencies which are typically in the THz range. Its overall magnitude increases with time as $E_{ek}(t)$ decays. Therefore, $E_{ik}(t)$ can be decomposed as a summation of a function $E_{ik,sm}(t)$, which increases smoothly with time, and a random thermal noise term $E_{ik,n}(t)$ (whose time average is zero), which is due to the finite size of the system. Including the internal kinetic energy or thermal noise leads to an overestimation of Q especially at higher temperatures as demonstrated in previous studies.[11–13] Hence, the internal kinetic energy (both the time-smooth part and the thermal noise) should be properly removed from the time-series data $E_k(t)$ to obtain $E_{ek}(t)$ and thereby to estimate Q accurately.

The step-by-step procedure involved in extracting the $E_{ek}(t)$ of a particular mode is shown in Fig. 2. First, $E_k(t)$ is passed through a Butterworth band-pass filter of known frequency [obtained from a fast Fourier transform (fft) of the energy time response] with a bandwidth of 10% of the center frequency. This effectively eliminates all the thermal noise and the contribution from modes other than that of interest. The output of this step is shown in Fig. 2(b). Note that this profile still includes $E_{ik,sm}(t)$. It can be observed that the minimum of each cycle in Fig. 2(b) increases with time, reflecting the increase in $E_{ik,sm}$ (conversion of E_{ek} to E_{ik}) in the system. The value of E_{ek} should, by definition, approach zero as the system approaches equilibrium. Since we are interested in $E_{ek}(t)$ only, this is brought down to zero by simply subtracting the minimum of each cycle. Figure 2(c) shows the actual decay of E_{ek} of the mechanical mode. It can be observed that the $E_{ek}(t)$ decays towards zero as the system reaches equilibrium after 6 ns. An exponential fit ($e^{-2\zeta\omega t}$) is subsequently performed, where ζ is the damping ratio and ω ($=2\pi f$) is the angular frequency, which can be obtained from the known frequency. The factor 2 in the exponent corresponds to fact that the frequency of the energy time response

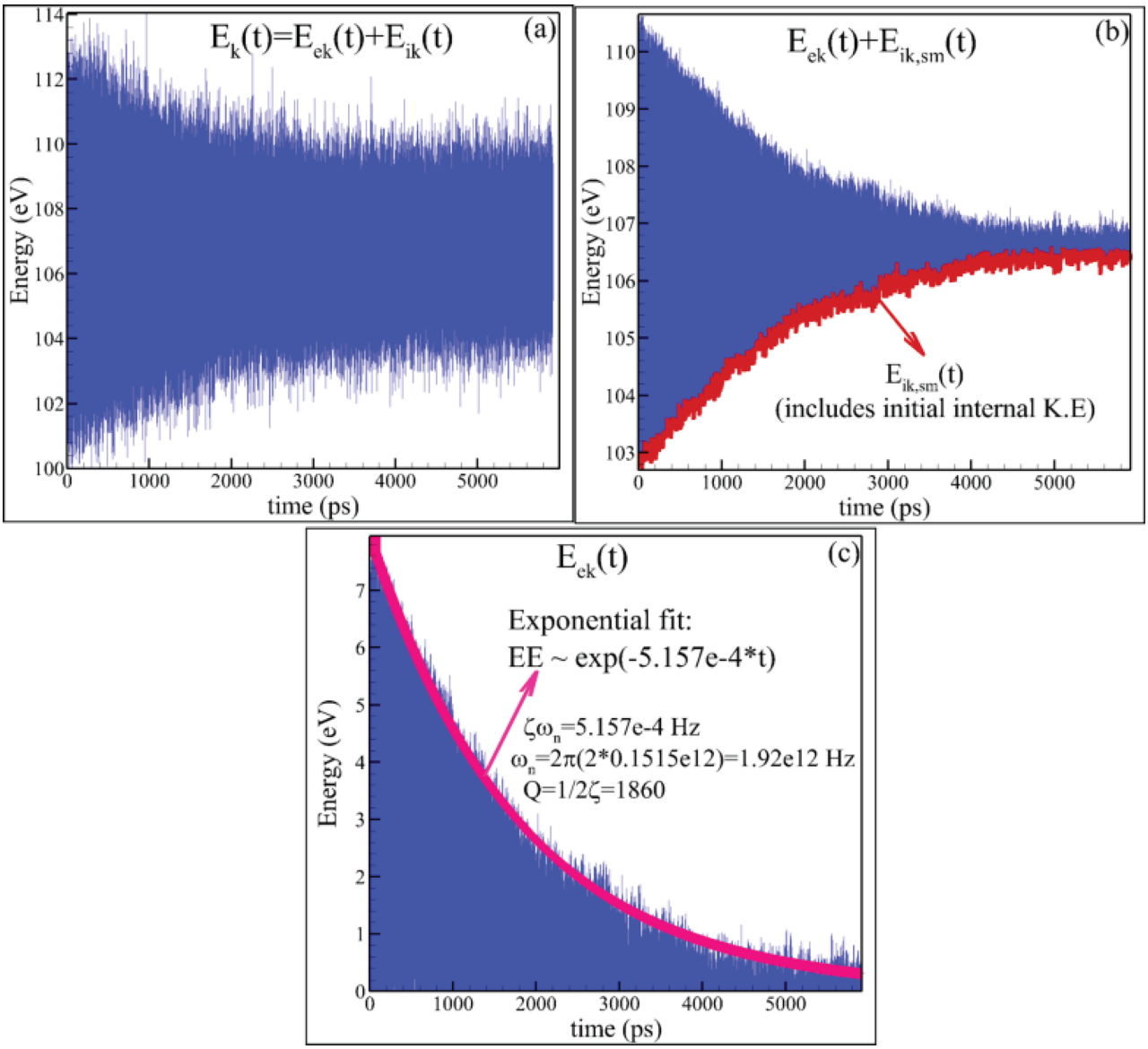

FIG. 2. Results of the various steps involved in calculating Q for a 16 nm long CNT at 300 K. (a) The variation of the total kinetic energy E_k with time post excitation. (b) The variation in the E_k value with time after the thermal noise has been filtered using a band-pass filter. (The line in red indicates the increase of internal energy with time.) (c) The variation of the external energy E_{ek} with time. (Also shown in red is the exponential fit to the decay of the E_{ek} amplitude, which is used to calculate Q.)

is twice that of the displacement. Q can then be directly estimated using the relationship ($Q = 1/2\zeta$). Note that calculating Q from the exponential decay temporal response is identical to the expression used in Ref. 11, as shown in the supporting information.[22] An alternative to using the temporal response of the energy is using the decay of the displacement of the COM to estimate the Q of the fundamental flexural mode,[14,17] which is labeled the RMS approach in Fig. 3. The authors have used this method previously to estimate the Q of cantilevered CNT resonators.[14] The displacement of the COM only captures the bulk motion of the CNT corresponding to a particular mode, as the thermal motion averages out to zero. This offers a significant advantage over the use of the temporal response of the kinetic (or potential) energy, which includes significant internal kinetic (thermal) energy at high temperatures. No further post-processing of data is necessary in this case and a direct fitting of the exponential curve ($e^{-\zeta\omega t}$) yields Q. Recently, this method has been used in experiments to determine the Q of silicon nanowires.[18,19] Figure 3 summarizes the temperature variation of Q calculated using each of the methods discussed above. It can be noted that the Q calculated from E_{ek} (green) and the RMS approach (red) are identical. Both methods conclude that Q varies inversely with T ($Q \sim T^{-1}$), which validates our aforementioned hypothesis that previous works have not accounted for the external energy accurately. It also demonstrates that including one or both of the components $E_{ik,sm}(t)$ and $E_{ik,n}(t)$ leads to non-classical temperature dependence, i.e., $Q \sim T^{-0.52}$ (blue) when all three components of E_k are utilized and $Q \sim T^{-0.63}$ (black) when the thermal noise ($E_{ik,n}$) is eliminated but the smoothed internal kinetic energy is not. Note that the Q in these two cases are also calculated

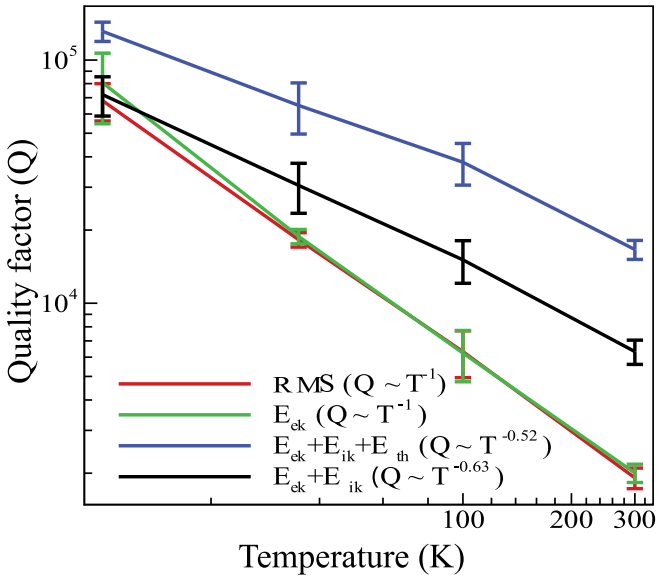

FIG. 3. The variation of Q factor with temperature using different computational methods.

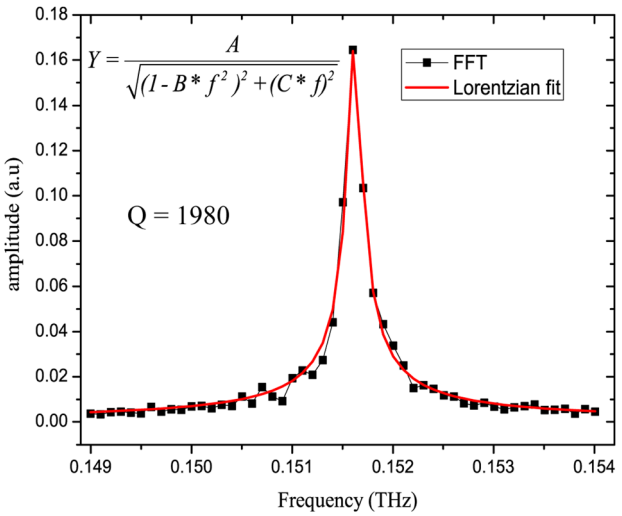

FIG. 4. FFT of the displacement of the COM at 300 K along with the Lorentzian fit.

from the exponential curve fit to the corresponding temporal response.

It is important to note that the RMS approach has its own limitations. It cannot capture axisymmetric motion like radial breathing modes (RBM) or even-order flexural modes in which the COM does not move. The filtering approach, on the other hand, is applicable as long as the mode has a distinct peak in the frequency response. Furthermore, although filter parameters like bandwidth and order may alter the magnitude of the output of the filter, the decay rate (and thereby Q) is relatively constant. For a mode like the fundamental flexural mode, the RMS approach is quite effective as it does not involve further post-processing, unlike the energy-based methods. It can be observed that at low temperatures, all methods yield similar Q values because of the low thermal noise and the relatively slow increase in internal energy. However, at high temperatures, the mismatch between the methods increases. The non-separation of the various energy contributions in the time response of energy leads to the weaker temperature dependence found in previous publications.[11–13] Also, we would like to point out that the energy ($E_{ek}(t)$ along with $E_{ik,n}(t)$) decays almost linearly rather than exponentially with time in Fig. 4 of Ref. 11; this is mainly because of the high Q (a result of low temperature) and less number of cycles used in that case. In such cases, using a linear or exponential fit will yield similar results. If the Q is low (at high temperatures) or a large number of cycles are used, exponential fitting as shown in Fig. 2(c) would be appropriate to predict Q accurately. The weak temperature dependence ($Q \sim T^{-0.36}$) reported by Huttel *et al.* in their experiments on SWCNT resonators at temperatures below 1 K may be either due to the presence of a substrate or due to quantum tunneling of two level systems which are dominant at low temperatures.[9] We would like to clarify that the agreement of this experimental result with the work of Jiang *et al.*[11] could be a mere coincidence as the experimental conditions ($T < 1$ K and $f = 293$ MHz) clearly indicate that the CNT is in a quantum mechanical ground state, the effects of which are impossible to capture in a classical MD simulation because of the high Debye temperature of CNTs (~500 K). The difference in the temperature range of experiments (0.01 K–1 K) and the MD simulation (1 K-293 K) is also to be noted in this regard. The inverse relationship of Q with T from the MD simulations suggests that the Q is in fact inversely proportional to the phonon population ($n = \hbar\omega/K_bT$), highlighting the role of the lattice anharmonicity (which causes phonon-phonon scattering) in the dissipative process. Also, note that applying a quantum correction to temperature leads to even stronger temperature dependence at low temperatures and it will gradually recover the classical limit as the system tends to the Debye temperature, as discussed in Refs. 14 and 20.

While the methods described above utilize the temporal response of the system, Q can also be calculated directly from the frequency response, as is frequently done in experiments. Consider the case where the first transverse mode is excited by applying a force at the center of the CNT. The Fourier transform of the displacement of the COM in the direction of the applied force exhibits a Lorentzian response, as shown in Fig. 4. By fitting a Lorentzian function (shown in the inset of Fig. 4) to this response, Q can be calculated directly based on the fitting parameters B and C (i.e., $Q = B/C$). It can be observed that the Q obtained from this method at 300 K is 1980, which agrees well with the value previously obtained by performing an exponential curve fit to the envelope (1860).

IV. FREQUENCY DEPENDENCE OF Q

In addition to understanding the temperature dependence of Q, it is interesting to determine the variation of Q for higher-order flexural modes. For this purpose, we have calculated the Q of the second and third flexural modes of the system employed earlier. The second and third modes are excited by applying forces at the locations of the antinodes of each mode shape, as shown in the insets of Fig. 5(a).

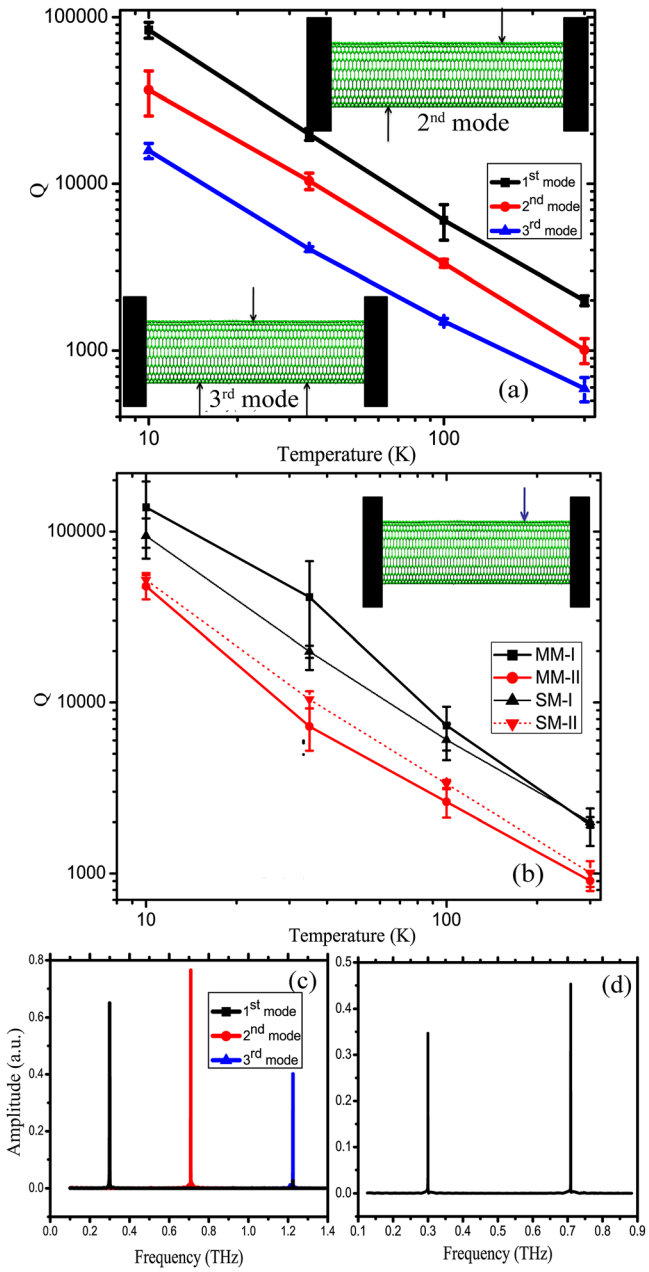

FIG. 5. (a) Variation of Q with temperature for different modes. (b) Variation of Q with temperature for the first (I) and second (II) modes under single mode (SM) and multi mode (MM) excitations. (c) Frequency response under single mode excitation [corresponding to (a)]. (d) Frequency response under multimode excitation [corresponding to (b)].

The frequency response of the kinetic energy is shown in Fig. 5(c), with the three peaks corresponding to the first three flexural modes. (The three colored peaks correspond to the three modes and are obtained from different simulations but are plotted together for illustration purposes.) We note that when the third mode is excited, there is a slight excitation of the first mode as well because of the similarity in the mode shape, though this is not obvious from the figure. The Q value is calculated using the filtering approach described earlier. The linear variation of Q with temperature for the first three modes is plotted in Fig. 5(a). Clearly, Q decreases with the increasing order of the mode (or the frequency) in an almost linear fashion. We have performed similar calculations on a 32 nm long CNT and obtained a similar frequency dependence. The reason for this linear frequency dependence can be explained using the relaxation times of transverse phonons in CNTs. The relaxation rate (Γ) increases quadratically at lower frequencies as a result of a three-phonon scattering process, as shown in Ref. 21. As a result, Q (also defined as ω/Γ) decreases linearly with ω. Hence, it may be advisable to operate the device at the first mode for maximum efficiency and sensitivity.

The fact that Q decreases linearly with ω ($=2\pi f$) also implies that the product fQ is almost constant for the CNT. This quantity is approximately equal to 3.5×10^{14} Hz at room temperature for the 16 nm long CNT. This product would decrease with the length of the CNT since the frequency decreases as $1/L^2$, whereas Q increases linearly with L.[14] It has been shown through experiments and simulations that applying tension can increase both f and the Q in doubly clamped nanowire and nanotube resonators, thereby enabling the increase of maximum attainable fQ for any device.[11,19]

We have also studied a case with multimode excitation where the first and second modes are excited simultaneously by applying a force at one quarter of the length from one of the ends as shown in Fig. 5(d). This plot also shows the excitation of the two modes in the frequency response. By applying suitable band-pass filters based on the known frequencies, we were able to distinguish the contribution of each of the modes separately. The magnitude of Q obtained in this manner is in fact equal to that of Q calculated earlier by exciting the same modes one at a time, as shown in Fig. 5(b). This is because the excited force is small enough that the response is linear. Hence, the two modes do not have any interaction (energy exchange) between them. In addition, the selection rule for three-phonon scattering in CNTs forbids any energy exchange between two flexural phonon modes with frequencies close to one another. This method demonstrates the advantage of employing a filter in scenarios where there are multiple modes excited in the system. For example, this method can be very relevant in the case of graphene nanoribbons where it is difficult to excite a single mode of vibration because of its thin, sheet-like structure.

V. CONCLUSIONS

To summarize, we have demonstrated that MD simulations enable the calculation of Q of a mechanical mode based on the CNT's time response as well as its frequency response. Using a band-pass filter, the external energy of the excited mechanical mode is accurately derived, leading to the conclusion that the temperature dependence of Q is in accordance with classical theory. By employing this method, we have shown that Q decreases with increasing order of the flexural mode. Also, we have shown that this filtering approach works well in the presence of multimode excitation, as long as the system operates within the linear regime. The general conclusions obtained from this work should hold true for other configurations, for example, cantilevers, which have fixed-free boundary conditions.

ACKNOWLEDGMENTS

A. Vallabhaneni acknowledges the financial support provided graciously by the Robert V. Adams endowment at Purdue University and would like to thank Nikhil Sheth for his assistance.

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[22]See supplementary material at http://dx.doi.org/10.1063/1.4754450 for the supporting information which includes the derivation of the expression showing the relation between external energy and quality factor.