Tunable, Broadband Nonlinear Nanomechanical Resonator

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ABSTRACT A nanomechanical resonator incorporating intrinsically geometric nonlinearity and operated in a highly nonlinear regime is modeled and developed. The nanoresonator is capable of extreme broadband resonance, with tunable resonance bandwidth up to many times its natural frequency. Its resonance bandwidth and drop frequency (the upper jump-down frequency) are found to be very sensitive to added mass and energy dissipation due to damping. We demonstrate a prototype nonlinear mechanical nanoresonator integrating a doubly clamped carbon nanotube and show its broadband resonance over tens of MHz (over 3 times its natural resonance frequency) and its sensitivity to femtogram added mass at room temperature.

KEYWORDS Nonlinear resonance, nanoresonator, broadband resonance, responsivity, carbon nanotube

Recent advances have seen the development of nanomechanical resonators operating in the linear regime that are capable of detecting extremely small physical quantities and even quantum interactions. However, the reduced device size reduces its dynamic range (down to the nanometer scale) for linear operation, which makes developing the required measurement system difficult and accordingly limits its sensitivity, especially under ambient and room-temperature environments.

The main element in most mechanical nanoresonators consists of a nanoscale mechanical cantilever or a nanoscale doubly clamped beam, which significantly reduces the effective mass of the resonance system. A general feature of such devices is that they operate predominantly in the linear regime and achieve high sensitivity to mass or charge through the realization of high quality-factor resonance at high frequency. Most noticeably, their recent development has allowed the sensing of mass down to the zeptogram (zg) level, for even a single molecule, and the transport of a single electron charge. Whereas the absolute magnitude of the involved resonance amplitude is small, the relative magnitude is actually quite significant when compared to the reduced device size. As a result, such nanoscale resonance systems can easily transition from linear resonance operation to a nonlinear one through a slight increase in its dynamic operating amplitude. The importance of nonlinearity in such nanomechanical resonance systems is thus gaining more recognition. For example, electrostatic interactions and coupled nanomechanical resonators were proposed for tuning the nonlinearity in nanoscale resonance systems; noise-enabled transitions in a nonlinear resonator were analyzed to improve the precision in measuring the linear resonance frequency and a homodyne measurement scheme for a nonlinear resonator was proposed for increasing the mass sensitivity and reducing the response time. In addition, the basins of attraction of stable attractors in the dynamics of a nanowire-based mechanical resonator were studied, and the nonlinear behavior of an embedded and a curved carbon nanotube was theoretically investigated. Such studies increasingly offer a new conceptual understanding and thus strategies to deal with and even exploit the increasingly prominent nonlinear behavior in the development of nanomechanical resonators.

Herein, we design an intrinsically nonlinear nanomechanical resonator defined by the inherent geometric nonlinearity that can be readily incorporated into practical device development, and we apply both theoretical modeling and experimental validation to demonstrate its tunability, its capacity for broadband resonance, and its sensitivity to added mass and to energy dissipation due to damping.

The intrinsic nonlinearity is simply introduced into the nanoscale resonance system through a geometric design as described in the following. Consider a fixed—fixed mechanical resonator employing a linearly elastic wire with negligible bending stiffness and no initial axial pretension. When driven transversely by a periodic excitation force applied locally to the middle of the wire, it will exhibit strong geometric nonlinearity and become an intrinsically (purely)

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nonlinear resonator (Figure 1A). In such a resonator, the force–displacement dependence is described by the relation 
\[ F = kx[1 - L(L^2 + x^2)^{-1/2}] \approx (k/2L^2)x^3 + O(x^5), \]
where \( F \) is the transverse point force applied to the middle of the wire, \( x \) is the transverse displacement in the middle of the wire, and \( L \) and \( k \) are the half-length and the effective axial spring constant of the wire, respectively. The total absence of a linear force–displacement dependence term (i.e., a term of the form \( kx \)) results in the realization of a geometrically nonlinear force–displacement dependence of pure cubic order. This resonator has no preferential resonance frequency, and its resonance response is broadband,\(^{26} \) which is conceptually different from typical linear mechanical resonators. Moreover, the apparent resonance frequency is completely tunable through the instantaneous energy of the system. If the bending effects are non-negligible or if an initial pretension exists in the wire, then a nonzero linear term in the previous force–displacement relation is included, giving rise to a preferential resonance frequency.

However, as long as this preferential frequency is sufficiently small compared to the frequency range of the nonlinear resonance dynamics, the previous conclusions still apply.

Thus, we proceed to analyze a doubly clamped Euler–Bernoulli beam having a foreign mass \((m_c)\) attached to its middle and excited transversely by an alternating center-concentrated force. Considering the geometric nonlinearity induced by axial tension during oscillation, the vibration of the beam is described by

\[
[pA + m_c\delta(x - L)]w_{tt} + (m\omega_o/Q)w_t + Elw_{xxxx} - \frac{(EA/4L)w_x}{w_t} \int_0^{2L} w_t^2 \, dx = F \cos \omega t \delta(x - L) \quad (1)
\]

where \( w(x, t) \) is the transverse displacement of the beam with \( x \) and \( t \) denoting the spatially and temporally independent variables, \( E \) and \( \rho \) are the Young’s modulus and mass density, \( A \) and \( L \) are the cross-sectional area and half length of the beam, \( I \) is the area moment of inertia of the beam, \( Q \) is the quality factor of the resonator in the linear dynamic regime, \( F \) is the excitation force applied to the middle of the beam, \( \omega = 2\pi f \) is the driving frequency, and \( \omega_o = (2\pi \omega_c) \) is the linearized natural resonance frequency of the beam. It is assumed that no initial axial tension exists when the beam is at rest, and shorthand notation for partial differentiation is used.

The transverse displacement of the beam can be approximately expressed as \( w(x, t) = \sum_{i=1}^N W_i(x) \phi_i(t) \), where \( W_i(x) \) is the \( i \)th mode shape of the linearized beam, \( \phi_i(t) \) is the corresponding \( i \)th modal amplitude, and \( N \) is the number of beam modes considered in the approximation. The leading modal amplitude, \( \phi_i(t) \), is then approximately governed by a Duffing equation obtained by discretizing eq 1 through a standard one-mode Galerkin approach\(^{27} \) (Supporting Information):

\[
(1 + M)i\ddot{\phi}_1 + \frac{\omega_o}{Q} \phi_1 + \omega_o^2 \phi_1 + \alpha \phi_1^3 = q \cos(\omega t) \quad (2)
\]

Here, \( M = \frac{[m_i/(2\rho AL)]W_i^2(L)}{(m_i/m_0)W_i^2(L)} \) is the ratio of the foreign mass to the overall mass of the beam multiplied by a factor due to the center-concentrated geometry of the foreign mass distribution (when the foreign mass is distributed evenly on the beam, \( M = m_c/m_0 \)); the amplitude of the driving force per unit mass in eq 2 is defined by \( q = W_i(L)F/(L \gamma) \); and the nonlinear coefficient is defined by

\[
\alpha = -\frac{E}{32\rho L^4} \int_0^{2L} W_i W''_i \, dx \int_0^{2L} (W'_i)^2 \, dx
\]

Following a harmonic balance approximation\(^{27} \) with a single frequency \( \omega \), we find that the response spectrum of
this Duffing oscillator forms a multivalued region when the oscillation amplitude is over a critical value as seen in the inset of Figure 1B. Specifically, there are two branches of stable resonances that are connected by a branch of unstable resonances. As the frequency is swept up, the resonance amplitude in the upper branch of stable resonances increases up to the maximum possible amplitude and then drops abruptly to a lower value as the forced motion makes a transition to the lower stable branch. The drop frequency, \( f_{\text{drop}} \), at which this jump phenomenon occurs is approximately determined by the intersection of the Duffing response spectrum with the free oscillation or the “backbone” curve, and its ratio to the linearized natural frequency is given by

\[
r_{\text{drop}} = \frac{f_{\text{drop}}}{f_o} = \left(1 + \frac{\sqrt{1 + M \Gamma}}{1 + M} \right)^{1/2}
\]

where \( \Gamma = \gamma((FQ)/(E))^2((2L)/D)^4((1)/(D^3)) \) and \( \gamma = 0.0305 \). From this equation, it is clear that the drop frequency of this nonlinear resonator strongly depends on the attached center mass and damping, besides the geometry of the beam and the applied excitation force. A similar computation can be performed for the reverse jump-up frequency during a decreasing frequency sweep; in that case, the dynamics follows a transition from the lower stable resonance branch to the upper.

We estimate the mass responsivity \( (R_m) \), defined as the shift in drop frequency with respect to the change in the added center mass, as

\[
R_m = \lim_{\Delta m_c \to 0} \frac{\Delta f_{\text{drop}}}{\Delta m_c} = -\frac{f_o}{2m_o} \left(1 - \frac{r_{\text{drop}}^2 - 1}{2r_{\text{drop}}^2 - 1}\right) W_1^2(L)
\]

where \( r_{\text{drop}} = \frac{f_{\text{drop}}}{f_o} \). Compared with a mass sensor based on a linear resonator for which the responsivity is \( -f_o/2m_o \), the nonlinear resonator utilizing the drop frequency as the measurant has a better responsivity by a factor of \( r_{\text{drop}}[1 - (r_{\text{drop}}^2 - 1)/(2r_{\text{drop}}^2 - 1)] \) when ignoring the term \( W_1^2(L) \) and if \( r_{\text{drop}} \geq 1.618 \). The mass responsivities for three representative doubly clamped beams with \( E = 100 \text{ GPa} \) and \( \rho = 2600 \text{ kg/m}^3 \) and a single-walled CNT beam with \( E = 1 \text{ TPa} \), for which the parameters are listed in the inset table, are plotted in Figure 2A as a function of the normalized frequency \( f_{\text{drop}}/f_o \). The value at \( f_{\text{drop}}/f_o = 1 \) indicates the responsivity of a linear resonator. It is apparent that the responsivity is enhanced not only by considering a nonlinear resonator with a smaller intrinsic mass and a higher resonance frequency but also by increasing the ratio of the drop frequency over the natural resonance frequency. This means that the performance of a mass sensor based on a nonlinear nanoresonator can be considerably raised by increasing its resonance bandwidth which, as we will show later, is practically tunable.

For a nonlinear resonator to have such intrinsically nonlinear behavior and a highly broadband resonance response, several parameters, including the quality factor, the size of the mechanical beam, and the driving force, are to be optimized to provide a larger value of \( \Gamma \) according to eq 3. Here, it is noted that the resonance bandwidth can be extended by simply increasing the excitation force while

\[\text{FIGURE 2. Sensing performance of a nonlinear nanoresonator to mass and energy dissipation due to damping. (A) Mass responsivities of four different doubly clamped beams as a function of the drop frequency/natural frequency ratio. (B) Shift in the drop frequency for a 1% change in the damping coefficient as a function of the drop frequency/natural frequency ratio. The inset table lists the parameters for the carbon nanotubes used in the calculation.}\]
keeping all other parameters of the resonator fixed. Figure 1B shows the tunability of the bandwidth up to 2 orders of magnitude by simply changing the excitation force applied to a nonlinear mechanical nanoresonator.

In addition, the drop frequency of the nonlinear nanoresonator is very sensitive to the magnitude of damping associated with the resonance system under various ambient conditions, according to eq 3. The damping responsivity of the drop frequency is estimated according to the change in the damping coefficient $\xi$, where $\xi = 1/(2Q)$:

$$
R_{\xi} = \lim_{\Delta /\!\!\!\!\!\!\xi \to 0} \frac{\Delta f_{\text{drop}}}{\Delta /\!\!\!\!\!\!\xi} = \frac{f_{g}}{\xi} r_{\text{drop}} \left( \frac{r_{\text{drop}}^{2} - 1}{2r_{\text{drop}}^{2} - 1} \right)
$$

The shift in drop frequency for a 1% change in the damping coefficient is plotted in Figure 2B and again shows the much enhanced sensitivity offered by the intrinsically nonlinear nanoresonator compared to that offered by the linear one.

We fabricated a nonlinear nanoresonator using a doubly clamped carbon nanotube (CNT), of which a scanning electron microscope (SEM) image is displayed in Figure 3A. The device was fabricated through micromachining and nanomanipulation. A silicon (100) wafer was coated with a 500-nm-thick silicon nitride layer followed by 1.5-$\mu$m-thick silicon dioxide. A thin Cr/Au layer was then sputter coated onto the silicon wafer and subsequently patterned through photolithography to form a three-electrode layout. This silicon wafer was back etched in KOH to make a thin membrane of silicon dioxide under the electrodes. The window was then milled with a focused ion beam to create three suspended electrodes. Three vertical platinum posts were fabricated onto these three electrodes through electron-beam-induced deposition. A high-quality multiwalled CNT produced with an arc discharge was then selected and manipulated inside an electron microscope and suspended between two of the platinum posts with both ends fixed with electron-beam-induced deposition of a small amount of platinum. The remaining platinum post was used as the driving electrode for applying a localized oscillating electric field to drive the oscillation of the CNT. The overall design of the device maximized the localization of the excitation force applied to the CNT beam (Supporting Information). According to the previous discussion, the localization of the applied force is necessary to create the strong geometric nonlinearity in the resonance system.

To acquire the response spectrum of the nanoresonator, the frequency of the applied ac driving voltage ($V_{ac}$) was swept up and then down while the oscillation amplitude in the middle of the CNT was measured from the acquired images in a SEM at room temperature and at a vacuum pressure of $\sim 10^{-6}$ Torr. To evaluate the effect of added mass on the dynamic behavior of the nanoresonator, a small amount of platinum was deposited on the middle of the CNT with electron-beam-induced deposition, and its mass was estimated from the measured dimension.

Figure 3B shows the acquired response spectrum for a nonlinear nanoresonator incorporating a CNT of $2L = \sim 6.2$ $\mu$m, $D = \sim 33$ nm driven with an ac signal of 10 V amplitude. The initiation of the oscillation started at around 4 MHz, near the natural resonance frequency of this doubly clamped CNT. The amplitude of the resonance oscillation increased continuously during the increasing frequency sweep up to 14.95 MHz, at which point the amplitude suddenly dropped to zero. This response closely resembled what was modeled previously for an intrinsically nonlinear nanoresonator and corresponded to a resonance bandwidth of over 10 MHz. During the ensuing decreasing frequency sweep, the resonator stayed mostly in a nonresonance state until reaching the neighborhood of the natural resonance.
frequencies of the CNT, where transitions back to resonance oscillations occurred. By fitting the obtained drop-jump and up-jump frequencies with the model prediction, the driving force was estimated to be $\sim 7$ pN and the Q factor of the system was $\sim 260$, which were in agreement with the estimate from an electrostatic analysis based on the experimental setup (Supporting Information) and the reported $Q$-factor values for typical CNT-based resonators, respectively.

The occurrence of multiple up-jump transitions during the decreasing frequency sweep appears to be due to the existence of multiple natural resonance frequencies in a multiwalled CNT and thus multiple modes of resonance. In theory, there are the same number of fundamental frequencies and resonance modes as the number of cylinders in a multiwalled CNT. In a recent computational study, it was shown that in the strongly nonlinear regime there can be coupling between multiple radial and axial modes of a double-walled CNT, with van der Waals forces provoking dynamic transitions between the modes of the inner and outer walls. Such strongly nonlinear modal interactions can be studied using asymptotic techniques in the context of coupled nonlinear oscillators.

The existence of multiple natural modes in this multiwalled CNT-based nonlinear resonator can also be revealed in an increasing frequency sweep when the driving force is reduced. Figure 3C shows the response spectrum acquired from the same resonator when the applied ac amplitude was reduced to 5 V. Two distinct resonance modes were excited in this case. The first mode appeared at around 4 MHz, and its drop jump occurred at 7.05 MHz. The second mode was then initiated right after the drop jump of the first mode and jumped down at 14.15 MHz. As shown previously, when the driving force was increased, it appeared that the first mode resonance became dominant and suppressed the initiation of the second mode in the increasing frequency sweep whereas in the decreasing frequency sweep, because there was no dominant mode, those modes were excited in the neighborhoods of their linearized resonance frequencies. Similar observations have been reported in coupled nonlinear resonators but not, until now, for a multiwalled CNT intentionally operated in a highly nonlinear regime.

The mass-sensing capability of the nonlinear nanoresonator is evaluated by adding a small platinum deposit to the middle of a suspended CNT, as shown in Figure 4. In this case, the CNT is $\sim 6.0$ $\mu$m long and $\sim 26$ nm in diameter. The added mass caused both a 2.0 MHz shift of the linearized natural frequency, approximately defined as the frequency where the resonance oscillation was initiated, and a more significant 7.4 MHz shift of the drop frequency. The added mass was estimated to be $\sim 7$ fg on the basis of the dimensions of the deposit measured from the acquired SEM images (Supporting Information). The corresponding mass responsivity calculated from the shift in the drop frequency ($R_{m, \text{nonlinear}} = 1.06$ Hz/fg) was 3.7 times that calculated from the linearized natural frequency ($R_{m, \text{linear}} = 0.29$ Hz/fg).

These mass responsivity values compare favorably with our model prediction from which $R_{m, \text{nonlinear}} = 2.18$ Hz/fg and $R_{m, \text{linear}} = 0.60$ Hz/fg. The magnitude of the shift in the drop frequency increases with the increase in added mass while in the meantime the bandwidth of the resonance decreases (Supporting Information).

This demonstration of a relatively simple nonlinear nanoresonator incorporating intrinsic geometric nonlinearity offers a model system for expanded studies of the nonlinear resonance behavior, which has been shown to be rich in physics and in opportunities for practical applications on the macroscale, now down to the nanoscale. In this study, we show a prototype as a mass sensor that can be applied to other types of high-sensitivity sensing applications. Compared with the sensing principles applied in nanoscale linear resonance systems, a nonlinear resonance system can exploit the instabilities intrinsically existing within the system that are very sensitive to external perturbation. The large oscillation amplitude and the sharp transition at these bifurcation points are all very favorable characteristics from the precision measurement point of view, which can potentially enhance the measurement sensitivity of a practical sensing system; the large oscillation amplitude implies less susceptibility of the resonance system to thermal noise, and a sharp transition allows for a narrow measurement bandwidth. However, the use of such a nonlinear system for high-sensitivity sensing applications may ultimately rely on our more detailed understanding of the robustness of such transitions related to the instability and the effect of external noise (thermal noise and stochastic perturbations). As shown in other studies, the complex dynamics of...
nonlinear systems and the instabilities associated with it are theoretically predictable and are robust enough for practical use in sensing applications.

Moreover, nonlinear resonance systems have recently been explored for more effective energy harvesting and more efficient mechanical damping applications because of their broadband resonance nature and unique characteristics favoring directional energy transfer in coupled systems. As demonstrated in this study, such broadband resonance behavior is preserved on the nanoscale and thus can be potentially exploited for nanoscale energy-harvesting and energy-transfer applications.

The design and demonstration of a simple nonlinear mechanical resonator, which operates on the nanoscale, expands the bandwidth of the resonance response, is tunable over a broad frequency range, and provides the inherent instabilities that can be exploited for sensing applications, offers new conceptual strategies for the development of nanoscale electromechanical devices. Such development is further facilitated by the inherent ease of realizing intrinsic geometric nonlinearity in a nanoscale resonator and can thus be readily integrated into the ongoing development of nanoscale electromechanical systems to extend their operation.

Supporting Information Available. Derivation of the drop frequency. Estimation of the applied driving force. Young’s modulus and the natural frequency of carbon nanotubes. Added mass produced with electron-beam-induced Pt deposition. Shift in the drop frequency and decrease in the bandwidth with increasing center mass. This material is available free of charge via the Internet at http://pubs.acs.org.

REFERENCES AND NOTES