Mode coupling bi-stability and spectral broadening in buckled carbon nanotube mechanical resonators

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Bi-stable mechanical resonators play a significant role in various applications, such as sensors, memory elements, quantum computing and mechanical parametric amplification. While carbon nanotube based resonators have been widely investigated as promising NEMS devices, a bi-stable carbon nanotube resonator has never been demonstrated. Here, we report a class of carbon nanotube resonators in which the nanotube is buckled upward. We show that a small upward buckling yields record electrical frequency tunability, whereas larger buckling can achieve Euler-Bernoulli bi-stability, the smallest mechanical resonator with two stable configurations to date. We believe that these recently-discovered carbon nanotube devices will open new avenues for realizing nano-sensors, mechanical memory elements and mechanical parametric amplifiers. Furthermore, we present a three-dimensional theoretical analysis revealing significant nonlinear coupling between the in-plane and out-of-plane static and dynamic modes of motion, and a unique three-dimensional Euler-Bernoulli snap-through transition. We utilize this coupling to provide a conclusive explanation for the low quality factor in carbon nanotube resonators at room temperature, key in understanding dissipation mechanisms at the nano scale.

A bi-stable system is the underlying operation principle of modern technology. It is the basic building block for computing, memories, and digital electronics. In MEMS technology, bi-stability is achieved by an arch-shaped beam with two symmetric possible configurations: either buckled up (or right) or down (or left). The transition between the two buckled configurations, named Euler–Bernoulli (EB) snap-through (ST) bi-stability, is a fascinating example of nonlinear behavior where the compression of the beam decreases its effective spring constant until it vanishes. The transition between the two configurations is controlled by an external force (usually electrostatic) and near this critical transition point, any small force perturbation results in a significant mechanical response, utilized for the realization of ultrasensitive force, acceleration and position sensors. Reducing the dimensions of a MEMS resonator to the nano scale improves its performance and enables observation of quantum effects that are not accessible via MEMS devices.

Carbon nanotube (CNT) based resonators have been widely investigated for sensing, signal processing, and quantum research. However, a bi-stable CNT resonator has never been demonstrated. In this work, we realize the EB snap-through buckling instability in novel suspended CNT resonators and investigate, both theoretically and experimentally, the static and the dynamic behavior of the system, and visualize the complete three-dimensional CNT motion. In fact, we present the first devices in which the CNT is initially buckled upward. This configuration enables unique out-of-plane static motion, which results in a completely different behavior than traditional CNT resonators. The realization of this type of device opens new avenues, some of which we report here: robust bi-stability for long-term endurance of the device, tenfold enhancement over the best electrical frequency tunability reported, and strong mode coupling explaining the anomalous dissipation of CNTs at room temperature.

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In devices exhibiting EB buckling, the geometric nonlinearity\textsuperscript{18,19} the constant pressure of $10^5$ Torr at room temperature, in agreement with the long-standing problem of low-quality factor of CNT resonators at room temperature, in agreement with the

discussion by the local-gate, and the detection is done by the classical resonances (in our case, from initial upward buckling to downward buckling) occurs through an abrupt mechanical transition, known as the snap-through Euler–Bernoulli buckling transition\textsuperscript{28,34}. This mechanical “jump” causes a change in the spring constant, which translates into a discontinuity in the resonance frequency (B $\rightarrow$ B’ in Fig. 1b, d). After the snap, decreasing the applied voltage, a second critical point is reached (D), resulting in an abrupt transition back to the original geometric configuration, known as the release (D $\rightarrow$ D’). The ST and release points may occur for different static loads (as is depicted in Fig. 1b), which then translates into a hysteric response, evidenced in both the DC conductance measurement (see supplementary text) and the resonance frequency measurement (Fig. 1d).

**Discussion**

We divide our devices into four different categories (Fig. 2 and S3). In the first category (device III, see Fig. 2a), the CNT is initially buckled downward ($b_0 > 0$), thus exhibiting the common behavior of CNT resonators which has been widely studied\textsuperscript{17,20}.

![Fig. 1 | Suspended CNT-based bi-stable NEMS device. a Schematic layout of the device, coordinates system and experimental setup for the resonance frequency measurement. Note that our coordinates system is such that the positive z direction points downward. Inset: SEM image of a typical device, displaying initial upward buckling. Scale bar=1$\mu$m. b Theoretical 2D model for the midpoint static deflection (red dot in a) as a function of the DC gate voltage. Our coordinates were chosen such that negative/positive values represent upward/downward curvature](https://doi.org/10.3390/s11467-022-33440-4)
The other categories, however, deal with the case of initial upward buckling ($b_0 < 0$), a regime never explored before in CNT resonators. These classes are characterized by the ratio between $b_0$ and the critical initial buckling, $b_c$, for which the ST instability is formed.

In the second group, the initial height is still minor compared to the critical value, meaning $|b_0| < |b_c|$. The resonance frequency gate dependence in this regime is characterized by a mild dip in the resonance frequency measurement (device IV, Fig. 2b). As $|b_0|$ increases, we enter the third category, in which the mild dip sharpens (devices V and I, Figs. 2c and 3c). The solid lines are the theoretical fitting of the initial shape of the CNT (solid arch), illustrating the different classes relative to the critical value $b_c$ (dashed arch). e–f Modeling the dynamic response of the same devices in (a–d), respectively. The solid lines are the theoretical fitting to the experimental data (black dots). The initial CNT heights extracted from the fit are $b_{0a} = 10 \text{ nm}$ for device III, $b_{0b} = 6 \text{ nm}$ for device IV, $b_{0c} = -38 \text{ nm}$ for device V, and $b_{0d} = -45 \text{ nm}$ for device VI, in agreement with our qualitative interpretation.

Unlike previous CNT bi-stable devices that relied only on pull-in\(^4\), which deteriorates after only several switches\(^28\), our ST transition is robust. The snap-through transition occurs in every DC gate sweep. Hence, during a resonance frequency measurement, it is repeated hundreds of times. For example, we extracted an average ST voltage from the measurement in Fig. 1d of $V_{\text{ST}} = -4.39 \text{ V}$ and a standard deviation of $\sqrt{\Delta V_{\text{ST}}} = 0.02 \text{ V}$. The relative deviation in the snap-through voltage ($\Delta V_{\text{ST}}/V_{\text{ST}} = 0.46\%$) in our CNT resonators is significantly smaller than in previous mentioned bi-stable nanobeams ($V_{\text{ST}} = 50 \text{ V}$, $\Delta V_{\text{ST}}/V_{\text{ST}} = >10\%$)\(^59\). This shows that the ST transition in our CNT resonators is sharp and requires significantly lower power consumption.

While these results are highly encouraging, they leave us with a puzzle. According to the 2D model (Fig. 1b), the ST transition occurs at extremum points, which simulate zero spring constant and hence zero resonance frequency ($f_0 = 0$). However, looking closer at Figs. 1d, 2d (of devices in the fourth category), we note that the “jump” does not take place at $f = 0$, but at 20–60 MHz above zero. In addition, for the down sweep in Fig. 1d (left), we observe a negative jump, which cannot be explained by the naïve model. To solve this puzzle, we had to realize that the model in Fig. 1b assumes that the CNT movement is constrained to the $x$–$z$ plane (in-plane), where in fact, there is a “hidden” out-of-plane component responsible for our observations.

In order to understand the origin of the different classes of devices and their characteristics, we model our devices as doubly clamped beams\(^{31,32,33}\). Since the CNT cross section is circular, we must consider the out-of-plane as well as the in-plane motion\(^31\). The out-of-plane motion and the electrostatic force towards the local-gate impose torque along the CNT axis\(^30\), forming a gate-dependent torsion. Thus, the tube develops a twist during its static motion towards the local-gate. As a result, we formulated a 3D theoretical model in which the Euler–Bernoulli equations of motion are solved considering all three degrees of freedom: in-plane ($\omega_z$), out-of-plane ($\omega_y$) and twist ($\omega_\theta$) (see supplementary text). We should emphasize the fundamental difference whether the CNT is slacked downward (type I) or buckled upward (types III–V), since only the latter can result in static out-of-plane motion. While many CNT studies took into account out-of-plane dynamics\(^39\) and initial deformation\(^32\), we are the first to consider out-of-plane initial and static deflection in the EB equations, such that significant static out-of-plane motion must evolve. This type of motion is new both for suspended CNTs analysis (traditionally slackened downward) as well as for buckled beams analysis, in which the $y$ (width) and $z$ (thickness) beam dimensions are usually different, restricting the motion to be solely in-plane (width $\gg$ thickness). Our simulations yielded excellent agreement with the experimental results (Figs. 2e–h and 3c, d) and allowed us to visualize the complete and unique CNT motion (Fig. 3e, f).

Regarding the puzzle raised earlier, as to why the “jump” occurs at a finite frequency, the theoretical analysis predicts that the lowest out-of-plane mode ($\omega_{\text{out}}$) is always lower than its in-plane counterpart ($\omega_{\text{in}}$, see Fig. 3c, d), while the torsional vibrational modes ($\omega_\theta$) are several orders of magnitude higher ($\omega_\theta / \omega_{\text{in}} \propto L/r$, where $r$ and $L$ are the tube radius and length, respectively). Therefore, the mode that reaches zero frequency and dictates the ST transition is actually the out-of-plane mode. Accordingly, the lowest in-plane mode, which is usually the only mode we can observe in our measurements, will automatically exhibit

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**Fig. 2** | Influence of initial buckling on the dynamic response. a–d Representative resonance frequency measurements of devices III–VI for the different categories, where a $b_0 > 0$, b $b_c < b_0 < 0$, c $b_0 < b_c < 0$, and d $b_0 < b_c < 0$. The insets are schematics of the initial shape of the CNT (solid arch), illustrating the different classes relative to the critical value $b_c$ (dashed arch). e–f Modeling the dynamic response of the same devices in (a–d), respectively. The solid lines are the theoretical fitting to the experimental data (black dots). The initial CNT heights extracted from the fit are e $b_{0a} = 10 \text{ nm}$ for device III, f $b_{0b} = 6 \text{ nm}$ for device IV, g $b_{0c} = -38 \text{ nm}$ for device V, and h $b_{0d} = -45 \text{ nm}$ for device VI, in agreement with our qualitative interpretation.
a “jump” in the resonance frequency even before reaching zero (device VII, Fig. 3b, d). We shall also note that without a twist, a negative jump (as in Fig. 1d left) cannot be accounted for. Occasionally, due to the rotational motion near the inflection point (representing the transition from up to down), we are able to detect evidence for the out-of-plane resonance frequency (marked by the arrows in Fig. 3a). Examining the CNT shape development (Fig. 3e, f) reveals how the in-plane and out-of-plane motions are strongly intertwined.

Broadening analysis

Lastly, with the experimental data at hand and having a model that reproduces the physical motion of the CNT, we address a longstanding question in the field of CNT resonators. The energy dissipation mechanisms of micro and nano resonators are an important characteristic of the device, commonly represented by their quality factor, $Q$, a quantity indicating how much energy dissipates in one period of oscillation. Unfortunately, the quality factor of CNT resonators at room temperature (~1000) is much lower than anticipated (~10000) when considering known dissipation mechanisms.

An important experimental study showed that spectral broadening due to geometrical symmetry breaking can account for the low $Q$ values at room temperature. However, theoretical simulations suggest another possible mechanism (“fluctuation broadening”), in which thermal fluctuations should cause strong coupling between the in-plane and out-of-plane modes of motion, resulting from the dynamic built-in strain along the CNT. This effect has never been proved experimentally due to the problematic estimation of the built-in strain in standard CNT resonators.

To study the dissipation mechanisms of the system, we calculate the quality factor according to $Q = f_0/\Delta f$, where $\Delta f$ is the full width at half maximum (FWHM) and $f_0$ is the resonance frequency (example of device IX in Fig. 4a). We compare the $\Delta f$ extracted from the data with a theoretical estimation of $\Delta f$ based on the fluctuation broadening theory (Fig. 4b, see supplementary text for details). In the supplementary text, we also examine the broadening due to symmetry breaking, which has been previously shown to explain the low $Q$ at room temperature. We show that symmetry breaking cannot account by itself for the spectral broadening evidenced in our measurements (supplementary text).

The broadening due to the in-plane and out-of-plane coupling is estimated according to equation S33 in ref. 20:

$$\sigma_f = \frac{1}{2\pi} \left| \frac{\partial \pi}{\partial \sigma_{\text{op}}} \right| \cdot \sigma_{\text{op}} \quad (1)$$

where $\sigma_{\text{op}}$ is the out-of-plane component of the CNT movement with thermal fluctuations given by $\sigma_{\text{op}} = \sigma_{\text{op}}/\omega_{\text{op}}$, in which $\omega_{\text{op}}$ represents the lowest out-of-plane resonance mode. The peak width is then estimated according to $\Delta f = 0.65\sigma_f$.

As evidenced in Fig. 4b, the fluctuation broadening theory yields remarkable compatibility with the experimental data. This is the most significant experimental evidence to date supporting this theory as the primary cause for low $Q$ in CNT resonators at room temperature. Moreover, we can show that the other modes’ fluctuations contribution to the broadening is negligible and that the lowest out-of-plane mode dominates (see supplementary information). To further examine this theory, we present a similar analysis on device IV at varying temperatures in the supplementary information (Fig. S9), also showcasing excellent agreement between the theoretical prediction and the experimental data.
the theoretical calculation based on Eq. 1. The excellent resonators (Fig. 2a) are the standard, the highest electrical frequency tunability is reached in type C (Fig. 2c). In addition, type D devices cating integrated mechanical-resonator circuits for advanced com-
surface. We speculate that this would open the possibility for fabri-
stable states the CNT is fully suspended with no physical contact to any
present a double-well potential CNT resonator, wherein both of the
broadening in our devices, in agreement with the

| Fig. 4 | Spectral broadening at room temperature. a Resonance frequency measurement of device IX in the third category, where \( b_1 < b_2 < 0 \). The yellow lines represent the frequency cross sections from which the data in (b) were extracted. Inset is an example taken at the yellow solid line. Blue dots are the data, orange solid line is a Lorentzian shape fit. b Quality factor (left) and spectral broadening (FWHM, right) extracted from (a) vs the static gate load (blue dots). The orange solid line is the theoretical calculation based on Eq. 1. The excellent fit implies that fluctuation broadening is the most significant mechanism to cause the low Q evidenced at room temperature.

To conclude, thanks to a high-quality fabrication process, we were able to identify that there exist several types of carbon nanotube-based resonators and that the main differentiating attribute is the shape of the as-fabricated buckled beam. We found that, unlike the previous common-knowledge, it is possible to fabricate a CNT-based resonator exhibiting Euler–Bernoulli snap-through bi-stability. The technological implication of the resonators classification is that while type A CNT resonators (Fig. 2a) are the standard, the highest electrical frequency tunability is reached in type C (Fig. 2c). In addition, type D devices present a double-well potential CNT resonator, wherein both of the stable states the CNT is fully suspended with no physical contact to any surface. We speculate that this would open the possibility for fabricating integrated mechanical-resonator circuits for advanced computing as well as fast, durable and energy-efficient memory elements. On the theory side, we found that the existence of the snap-through bi-stability is associated with all three degrees of freedom of the CNT. Having a good experiment-model agreement, we also addressed the issue of an anomalously low-quality factor at room temperature and showed that nonlinear mode coupling accounts well for the spectral broadening in our devices, in agreement with the fluctuation broadening theory.

**Methods**

**Fabrication**

The geometric structure of our devices is presented in Fig. 1a. As a substrate, we used a silicon wafer with a layer of 500 nm silicon oxide on top. All patterning stages were done by standard photolithography or electron beam lithography procedures. We evaporated Cr/Pt 5/35 nm for the source and drain (SD) electrodes, after which we etched the SiO₂ using buffer oxide etchant (BOE) to create the local-gate trench (see supplementary text for details). Then, another step of Cr/Pt 5/35 nm evaporation was performed to create the self-aligned local-gate. Finally, we deposited a patterned ferritin catalyst near the SD electrodes for the CNT growth. The CNTs were grown by chemical vapor deposition (CVD) under a constant flow of 0.5/0.5 SLPM H₂/CH₄ at 900 °C for 20 min, utilizing the fast heating growth technique.

**Resonance frequency measurements**

The resonance frequency measurements were conducted using the mixing technique, presented schematically in Fig. 1a. The actuation of the CNT is done by applying an electrostatic force from the local-gate:

\[
F = \frac{1}{2} \frac{\partial C_g}{\partial z} V_{sd}^2 = \frac{1}{2} \frac{\partial C_g}{\partial z} (V_{gdc}^+ + V_{gdc}^- \cos(\omega t))^2
\]

where \( C_g(z) \) is the capacitance between the CNT and the local-gate electrode, \( V_{sd} \) is the local-gate voltage, which is a combination of a DC voltage \( V_{gdc} \) superimposed with a driving voltage \( V_{gdc}^+ \) excited at frequency \( \omega \).

For the mixing, we apply a second driving voltage between the SD electrodes, \( V_{sd} \):

\[
V_{sd} = V_{gdc}^+ \cos(\omega t + D\omega t)
\]

where \( V_{sd}^+ \) is the driving voltage, and \( D\omega t \) is the offset frequency (in our experiments, a few kHz). Utilizing the fact that the CNT serves as a mixer, we can actuate it at \( \omega \) and detect its resonance frequencies through the current at the offset frequency \( D\omega \):
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Author contributions

S.R. and T.T. have equal contributions to this work, took part in device fabrication, and performed the experiments and analysis. M.S. developed the fabrication process. S.S. assisted in the theoretical modeling and performed finite element simulations. S.R., T.T., and Y.E.Y. designed the experiment and wrote the manuscript, with input from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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