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Sources of nonlinearity of a driven carbon nanotube electromechanical resonator

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Nanoelectromechanical resonators provide an ideal platform for investigating the interplay between electron transport and nonlinear mechanical motion. Externally driven suspended carbon nanotubes, containing an electrostatically defined quantum dot, are especially promising. These devices possess two main sources of nonlinearity: the electromechanical coupling and the intrinsic contributions of the resonator that induce a Duffing-like nonlinear behavior. In this work, we observe the interplay between the two sources across different driving regimes. The main nonlinear feature we observe is the emergence of archlike resonances in the electronic transport when the resonator is strongly driven. We show that our model is in good agreement with our experimental electron transport measurements on a suspended carbon nanotube. This characterization paves the way for the exploration of nonlinear phenomena using mesoscopic electromechanical resonators.

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I. INTRODUCTION

Nanomechanical resonators have emerged as a unique platform for the exploration of nonlinear dynamics. They offer insights into various nonlinear phenomena, such as spontaneous self-oscillations [1–3] and bistable states of mechanical motion [4,5]. For this reason, they have been suggested for wide range of applications across different scientific fields, ranging from thermodynamics [6] to chaos [7] and metrology [8].

One type of resonator of particular interest is suspended carbon nanotube (CNT) due to their large zero-point motion [9,10] and large quality factors [11]. Here, we study a suspended CNT device that is externally driven (see Fig. 1). It is possible to electrostatically define a quantum dot (QD) along the CNT, and its electronic occupation is strongly affected by the motion of the CNT [12–15]. In turn, the motion of the CNT is altered by the electronic tunneling through the QD. Indeed, it has recently been reported that the coupling between

electron tunneling and mechanical motion is in the ultrastrong regime, resulting in strong backaction [16,17].

In the platform studied here, there are two distinct sources of nonlinearities affecting the mechanical motion: (1) the backaction due to the electromechanical coupling and (2) intrinsic mechanical nonlinearities. The contributions of each source depend strongly on the driving power [18–20]. Typically, at low driving power, source (1) dominates [18,21,22]. Meanwhile, source (2) starts to play a more predominant role at higher driving powers. Signatures of this second nonlinearity have been previously observed experimentally presenting as a Duffing-type response [18,23,24]. Although a lot of efforts have been made to model each nonlinear source independently [17,18,21,23,25–34], to our knowledge, the interplay between these two effects has not been yet explained.

In this work, we unify these nonlinear contributions within a single microscopic framework by deriving a theoretical model, from first principles, valid across all external driving regimes. We are specifically interested in the response of the system in the nonequilibrium steady state. Here, we fully characterize the contributions of the nonlinear sources and observe their features in the amplitude of the CNT and the time-averaged electronic current through the device. First, we study the case where a single transport channel plays a role in the electronic transport. When the CNT is weakly driven, source (1) is predominant, and we recover the typical frequency softening due to the QD-CNT coupling [18,21]. As the driving of the CNT increases and source (2) starts to become

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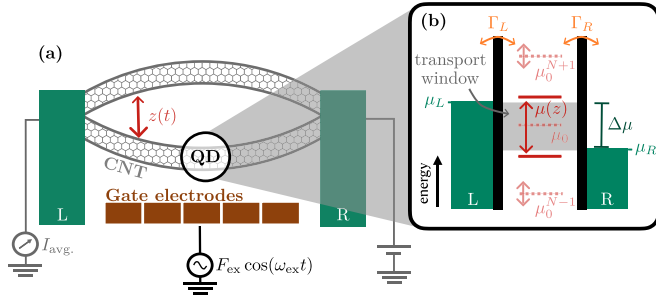


FIG. 1. (a) Schematic of the platform: a CNT is suspended between the left (L) and right (R) metal electrodes that act as fermionic reservoirs for the embedded QD. The QD is electrostatically defined via the gate electrodes. Furthermore, in one of the gates, an external AC drive with force F_{ex} and frequency ω_{ex} excites the displacement z of the CNT. Application of a bias voltage $\Delta\mu$ between the L/R reservoir drives a current I_{avg} through the device. (b) Sketch of the electrochemical potential structure of the embedded QD characterized by electrochemical potentials μ_0^N . Due to the electromechanical coupling, the electrochemical potential acquires a displacement dependence $\mu(z)$. We include two additional electrochemical potentials of the QD, μ_0^{N-1} and μ_0^{N+1} , which become relevant when comparing with experimental results in Sec. III. The L and R fermionic reservoirs have electrochemical potentials μ_L and μ_R , respectively, which enable a transport window where electrons in the QD can hop from L to R or vice versa with tunneling rates Γ_L and Γ_R .

dominant, archlike resonance structures appear in the electronic current. This appearance is the main feature of our model. To compare our theoretical findings with the experimental results presented here, we generalize our model to account for multiple transport channels. We find that our theoretical model agrees well with experimental results. This characterization is essential to pave the way for the exploration of fundamental experiments, such as observing signatures of quantum-to-classical transition at the nanoscale [35].

II. SINGLE ELECTROCHEMICAL POTENTIAL MODEL

A. Microscopic model

The system is composed of an externally driven suspended CNT that is free to vibrate with displacement z [see Fig. 1(a)]. A QD is electrostatically defined along the CNT such that it is coupled to its mechanical motion [16,18,21,22]. For now, we consider this QD to have a single transport channel, i.e., an electrochemical potential μ_0 within the bias window. In Sec. III, we will generalize the single electrochemical potential model to the multiple level case, as depicted in Fig. 1(b).

The full system is described by the spinless Anderson-Holstein model, with additional mechanical terms to model the intrinsic nonlinearities of the resonator and the external driving. We have

$$\hat{H} = \hat{H}_{\text{AH}} + \hat{H}_{\text{duff}} + \hat{H}_{\text{drive}}, \quad (1)$$

where first term constitutes the standard Anderson-Holstein model [23,36–38]

$$\hat{H}_{\text{AH}} = \hat{H}_{\text{QD}} + \hat{H}_{\text{leads}} + \hat{H}_{\text{T}} + \hat{H}_{\text{CNT}} + \hat{H}_{\text{CNT-QD}}. \quad (2)$$

Here, the first term is the Hamiltonian of the QD, $\hat{H}_{\text{QD}} = \mu_0 \hat{n}$, with occupation $\hat{n} = \hat{d}^\dagger \hat{d}$, where $\hat{d}^\dagger$ ($\hat{d}$) is the creation

(annihilation) operator of the dot. Electronic reservoirs are located to the left (L) and right (R) of the dot and are governed by the Hamiltonian $\hat{H}_{\text{leads}} = \sum_v \epsilon_v \hat{c}_v^\dagger \hat{c}_v$, where ϵ_v are the energies of the electrons in reservoir v and the operators $\hat{c}_v^\dagger$ ($\hat{c}_v$) are their respective creation (annihilation) operators. Throughout this work, we consider these reservoirs to always be in local thermal equilibrium. Equation (2) further models single-electron transport between the dot and the reservoirs by the tunneling Hamiltonian $\hat{H}_{\text{T}} = \sum_v t_v \hat{c}_v^\dagger \hat{d} + \text{H.c.}$, where t_v is the coupling constant related to the tunneling rate Γ_v . We take energy-independent rates, also known as the wideband limit [39–41]. The third term of Eq. (2) corresponds to the linear dynamics of the CNT motion, $\hat{H}_{\text{CNT}} = \frac{1}{2m} \hat{p}^2 + \frac{1}{2} m \omega^2 \hat{z}^2$, where $\hat{p}$ and $\hat{z}$ are the momentum and displacement operators of the mechanics, respectively, m is the mass, and ω its natural frequency. Finally, the last term of Eq. (2) is the linearized coupling between the mechanics and the electronic transport, given by [16,37,38]

$$\hat{H}_{\text{CNT-QD}} = \hbar \omega \lambda \frac{\hat{z}}{z_{\text{ZPM}}} \hat{n}, \quad (3)$$

where the mechanical zero-point motion is $z_{\text{ZPM}} = \sqrt{\hbar/2m\omega}$ and the unit-free parameter λ quantifies the strength of this electromechanical coupling. If $\lambda \ll 1$, the QD and the CNT motion are weakly coupled and thus can be treated perturbatively. On the contrary, if $\lambda \gtrsim 1$, we are in the ultrastrong coupling regime, where the electromechanical interaction significantly affects both the QD and the CNT motion. In what follows, we will focus on the ultrastrong case, in agreement with recent experimental characterizations of these devices [16,17]. Due to the coupling between the mechanics and the dot, the electrochemical potential of the dot depends on the displacement $\hat{z}$. We take here a first-order expansion in displacement as [16]

$$\mu(\hat{z}) = \mu_0 + \hbar \omega \lambda \frac{\hat{z}}{z_{\text{ZPM}}}. \quad (4)$$

An additional term in the total Hamiltonian [see Eq. (1)] is included to go beyond the standard Anderson-Holstein model and capture the intrinsic mechanical nonlinearity of the CNT, which we here take as a Duffing term [20,22]

$$\hat{H}_{\text{duff}} = \frac{\beta}{4} \left(\frac{\hat{z}}{z_{\text{ZPM}}} \right)^4, \quad (5)$$

where β quantifies the strength of these nonlinearities.

Finally, the last term of Eq. (1) describes the external AC drive with force F_{ex} and frequency ω_{ex} ,

$$\hat{H}_{\text{drive}} = F_{\text{ex}} \cos(\omega_{\text{ex}} t) \hat{z}. \quad (6)$$

Contrary to previous works [17,18,21,23,25–34], with the model presented here [Eq. (1)], we are able to simultaneously capture the two physical nonlinearities of the system: (1) due to the electromechanical coupling [Eq. (3)] and (2) from the intrinsic mechanical nonlinearities [Eq. (5)].

For the remainder of this article, we will focus on the nonequilibrium steady state of the system. For this, we consider the *quasiadiabatic limit*, where the oscillations of the resonator are slow compared to the electron tunneling, i.e., $\omega \ll \Gamma_L, \Gamma_R$. In this regime, a semiclassical model can be employed [16,37,42], which is given as follows. First, we

neglect the higher-order moments of the oscillator and consider only the average position $\langle \hat{z} \rangle = z(t)$. Thus, from the full Hamiltonian detailed in Eq. (1), the displacement with respect to the zero-point motion $x(t) = z(t)/z_{\text{ZPM}}$ obeys a classical equation of motion [43]

$$\ddot{x} + \delta \dot{x} + \omega^2 [x + 2\lambda n(x)] + \epsilon x^3 = f_{\text{ex}} \cos(\omega_{\text{ex}} t), \quad (7)$$

where $\epsilon = \beta/(mz_{\text{ZPM}}^2)$ is the normalized Duffing parameter and $f_{\text{ex}} = F_{\text{ex}}/(mz_{\text{ZPM}})$ is the normalized external driving (see Appendix A for a detailed derivation). Here, we have also included a damping term $\delta \dot{x}$ to properly account for the intrinsic damping of the CNT motion. Note that in Eq. (7) we retain the combined effect of both the intrinsic and transport-induced nonlinearities, which, as we will see, are necessary to properly capture the observed behavior of the CNT across all driving strengths. In Eq. (7), n is the average population of the dot, i.e., $\langle \hat{n} \rangle = n(t)$. We will assume that the displacement-dot correlations can be neglected, which is $\langle \hat{z} \hat{n} \rangle = \langle \hat{z} \rangle \langle \hat{n} \rangle$, similarly to previous works [16]. Typical CNT experiments operate in the lifetime broadened regime, where the electronic tunneling rates Γ_v are much larger than the fermionic reservoir temperatures $k_B T_v$. In such case, QD rate equations are not strictly valid and a more sophisticated approach is needed. We follow Refs. [39,41], where it is shown that under the wideband limit and $T_L, T_R \simeq 0$ assumptions, the average population of the dot is given by

$$n(x) = \frac{1}{2} + \frac{\Gamma_R}{\pi \Gamma_{\text{tot}}} \left[\arctan(\gamma_R(x)) + \frac{\Gamma_L}{\Gamma_R} \arctan(\gamma_L(x)) \right], \quad (8)$$

with $\gamma_v(x) = 2(\mu_v - \mu(x))/(\hbar \Gamma_{\text{tot}})$ and $\Gamma_{\text{tot}} = \Gamma_L + \Gamma_R$ is the total tunneling rate. We note that due to the explicit dependence of the QD electrochemical potential on x , an implicit time dependence is also acquired via $x(t)$.

We are now ready to characterize the nonequilibrium steady-state response of the QD and CNT motion. In Sec. II B, we will analyze the displacement of the CNT for different driving regimes. Then, in Sec. II C, we study the response of the QD population by computing the electron current flowing through the device.

B. Nonequilibrium steady-state response

In this section, we study the nonequilibrium steady-state displacement of the CNT. For this, we simultaneously solve Eqs. (7) and (8) (see Appendix B for details).

In Fig. 2(a), we plot the bare population of the QD, i.e., setting $\lambda = 0$, to characterize the regions of μ_0 where electronic transport is enabled (highlighted in gray). Then, taking the electronic backaction into account, i.e., $\lambda \neq 0$, we turn our attention to the nonequilibrium steady-state displacement of the CNT, x^{NESS} . This is plotted in Figs. 2(b)–2(d), as a function of the driving frequency ω_{ex} and the bare QD energy μ_0 for different driving forces.

Figure 2(b) corresponds to the weak driving regime, where ω_{ex} is swept from low to high values. If we trace a line cut in a region where μ_0 is outside the transport window, labeled as (1), we observe that the CNT responds as a driven harmonic oscillator [see panel (b-1)]. As μ_0 approaches the transport window, situation labeled as (2), the resonance frequency

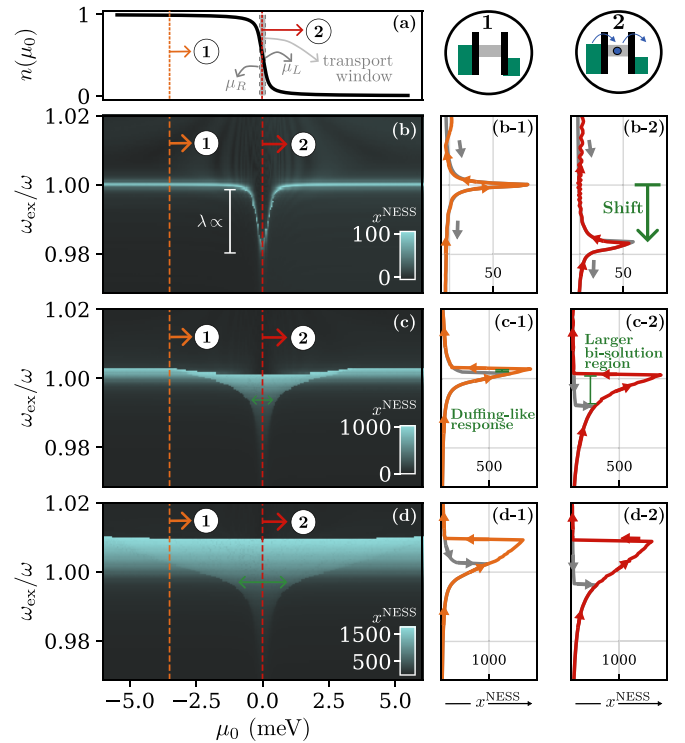


FIG. 2. (a) QD population as a function of the charging energy μ_0 . The shaded gray region identifies the values of μ_0 where electronic transport is enabled. For simplicity, the CNT motion's backaction is not taken into account, i.e., $\lambda = 0$. Displacement of the CNT motion with respect to its zero-point motion $x^{\text{NESS}} = z^{\text{NESS}}/z_{\text{ZPM}}$ in the nonequilibrium steady state as a function of both the external driving frequency ω_{ex} and the bare QD energy μ_0 for increasing normalized external driving forces as (b) $f_{\text{ex}}/\omega^2 = 0.08$, (c) $f_{\text{ex}}/\omega^2 = 0.8$, and (d) $f_{\text{ex}}/\omega^2 = 1.6$. In these panels, the driving frequency ω_{ex} swept from low to high values. (b-1)–(d-1). Frequency response of the CNT for a constant QD energy of $\mu_0 = -3.5$ meV (where no transport is present) highlighted as (1) in panels (b)–(d). In orange, the ω_{ex} is swept from low to high values, while the opposite sweeping direction is presented in gray. Similarly panels (b-2)–(d-2) also showcase the frequency response of the CNT, but for $\mu_0 = 0$ meV (where transport is enabled) highlighted with (2) in panels (b)–(d). In this case, ω_{ex} is swept from low to high values in red, while the opposite sweeping direction is again presented in gray. Parameters: $\delta/\omega = 9.4e-9$, $\epsilon/\omega^2 = 8e-4$, $\lambda = 3$, $\mu_L = -\mu_R = 0.1$ meV, and $\Gamma_L/2\pi = \Gamma_R/2\pi = 30$ GHz.

presents a softening dip shifting the resonance peak to lower ω_{ex} [see panel (b-2)]. The height of this dip depends on the coupling ratio λ . At higher λ , nonlinearities arise due to the transport backaction (see also Ref. [22]).

As the external driving force is increased [Fig. 2(c)], the CNT maximum displacement increases and the frequency response broadens. At an intermediate driving force, both sources of nonlinearities play a role. Inside the transport regime, the softening dip is still present but wider (highlighted in green). If we again make a line cut for values of μ_0 outside [panel (c-1)] and inside [(c-2)] the transport window, we note that the frequency response resembles a typical Duffing oscillator with a sharp drop for $\omega_{\text{ex}} > \omega$ [44]. Furthermore, hysteresis is present in both situations: the response

is different as a function of the external frequency direction sweep. This is a consequence of the bistability caused by the nonlinear Duffing term. Note that the region of bistability is increased when electronic transport is enabled compared to when it is not. This is an important observation as it is a key feature for some applications such as to observe quantum-to-classical transitions [35].

Finally, in Fig. 2(d), we present the strongly driven regime. Here, the intrinsic nonlinearities of the CNT motion dominate; thus, the Duffing-like response is accentuated, though the broadened frequency dip due to the electronic backaction is still present (highlighted in green).

Now that we have a good understanding of the different responses of the CNT motion for different driving regimes, we move on to study in the next section the signatures of the QD response in the electronic current flowing through the device that is experimentally accessible.

C. Measurable observable: the electronic current

In this section, we focus on the nonequilibrium steady-state response of the QD. Instead of analyzing the dot population [Eq. (8)], we compute the electronic transport through the device that allows us to easily compare with experiments. Again, following Refs. [39,41], the instantaneous current through the device is

$$I(x) = \frac{e}{\pi} \frac{\Gamma_L \Gamma_R}{\Gamma_{\text{tot}}} [\arctan(\gamma_L(x)) - \arctan(\gamma_R(x))], \quad (9)$$

where the parameters are the same as presented in Eq. (8). In experiments, what is measured is the averaged current over various oscillation periods T in the nonequilibrium steady state. Therefore, for the remainder of the work, we will focus on the average current

$$I_{\text{avg}} = \frac{1}{T} \lim_{\tau \rightarrow \infty} \int_{\tau}^{\tau+T} dt I(x(t)), \quad (10)$$

with $x(t)$ given by the nonequilibrium steady-state solution of Eq. (7). In Fig. 3, we plot the average current as a function of the driving frequency ω_{ex} and the bare QD energy μ_0 for different driving forces. In the main plots, ω_{ex} is swept from low to high values while the insets show the opposite case. In the weak driving regime, corresponding to panel (a), away from the mechanical resonance, Coulomb peaks are observed [45]. When the driving frequency approaches the natural frequency of the resonator, $\omega_{\text{ex}} \simeq \omega$, a softening dip in the current is present, as previously observed in multiple experiments [16,18,21,46]. The behavior is in accordance with the resonance frequency softening dip of the CNT motion [see Fig. 2(b)]. The reason for this decrease of I_{avg} is due to the fact that the backaction of the CNT motion displaces the electrochemical potential of the QD outside of the transport window (see right panel), and thus on average the current is reduced.

Next, we increase the driving force [see Fig. 3(b)]. We reproduce experimental observations made in Refs. [18,24], where the frequency dip is broadened with a sharp current change around $\omega_{\text{ex}}/\omega \simeq 1.05$. Similar to the displacement plot [Fig. 2(c)], the contribution of both sources of nonlinearities play a role. For this driving regime, the CNT displacement

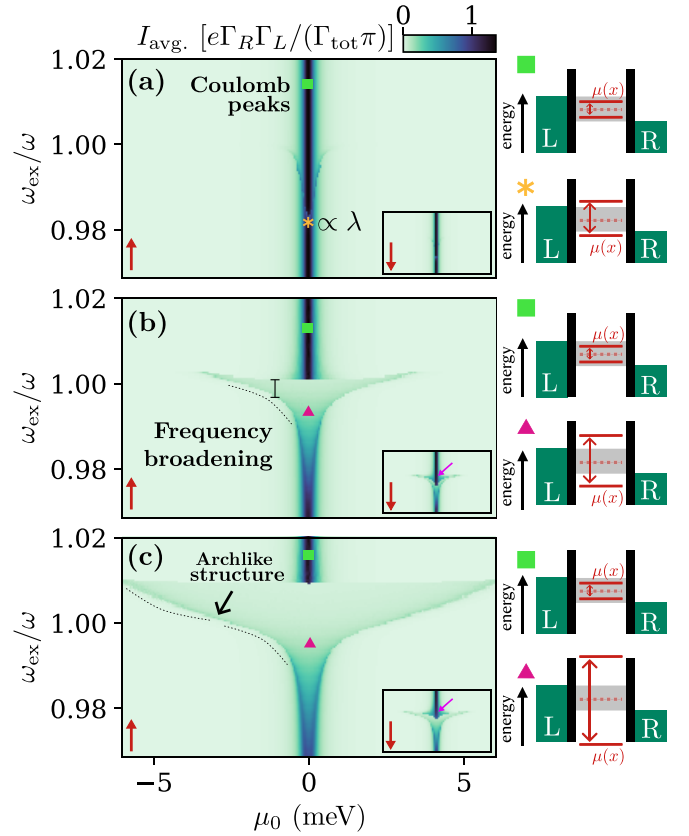


FIG. 3. Average electronic current I_{avg} for three increasing forces labeled as (a) $f_{\text{ex}}/\omega^2 = 0.08$, (b) $f_{\text{ex}}/\omega^2 = 0.8$, and (c) $f_{\text{ex}}/\omega^2 = 1.6$ as a function of the driving frequency ω_{ex} and QD bare electrochemical potential μ_0 . In the main panel, ω_{ex} is swept from low to high values, while in the inset ω_{ex} is swept in the opposite direction as indicated by red arrows. In panels (b) and (c), we have added dashed black lines to aid visualization of the archlike features defined in the main text. The right panels show an energy sketch of the QD, when ω_{ex} is far from the CNT's bare frequency ω (green square), and when ω_{ex} is close to ω , in which case μ is strongly affected by the displacement of the CNT (orange asterisk and magenta triangle). Parameters: $\delta/\omega = 9.4e-9$, $\epsilon/\omega^2 = 8e-4$, $\lambda = 3$, $\mu_L = -\mu_R = 0.1$ meV, $\Gamma_L/2\pi = \Gamma_R/2\pi = 30$ GHz, and $T = 4\pi/\omega_{\text{ex}}$.

presented a hysteresis behavior, which is also observed in I_{avg} . This is clearly seen by comparing the main panels of Fig. 3 to the insets, where ω_{ex} is swept in the opposite direction. Indeed, when ω_{ex} is swept from high to low values, instead of a sharp stop at $\omega_{\text{ex}}/\omega \simeq 1.05$, a small dip (marked by a magenta arrow) is present, which persists for higher driving forces.

Finally, in the strong driving regime [Fig. 3(c)], the transport response presents a very distinct feature. The broadening of the frequency outside the transport window is distorted: contrary to the intermediate driving case in Fig. 3(b), the curvature changes sign from negative to positive at the location indicated by the black arrow. To aid visualization, we have added dashed black lines highlighting this transition. We refer to this feature as an archlike shape. Inside this resonance, the current is suppressed. This is due to the effective electrochemical potential of the QD being “kicked out” of the transport window (see right panels). Experimental observations of these

features have been reported previously in Ref. [23]. Comparing with Fig. 2(d), we conclude that the archlike resonance arises due to the intrinsic nonlinearities modeled by $\hat{H}_{\text{duff}}$ in Eq. (1). The appearance of these archlike resonance composes a main feature of our model.

In the following section, we generalize our model to multiple electrochemical potentials of the QD entering the bias window and compare with experimental results.

III. GENERALIZATION TO MULTIPLE QD ELECTROCHEMICAL POTENTIALS AND EXPERIMENTAL RESULTS

Up to now, we have studied in detail the nonequilibrium steady-state response given by the model detailed in Sec. II for different external driving forces. We identified the key features both in the displacement of the CNT (Sec. IIB) and the current for a single electrochemical potential (Sec. IIC). In this section, we extend the model to take into account multiple electrochemical potentials in the QD: $\{\mu_0^i\}_{i=1, \dots, N-1, N, N+1, N+2, \dots}$ [see Fig. 1(b)]. As we will see later, this generalization is required to properly reproduce our experiments. If the displacement of the resonator (with respect to its zero-point motion) is small, the QD energy μ does not present strong deviations with respect to μ_0 . Thus, it is safe to say that the electronic transport can only be due to a single electrochemical potential entering the transport window. On the contrary, as the driving force increases, so does the displacement, and therefore the change in μ can be large enough that different charged states can contribute to the current.

To extend our model, we first consider that each electrochemical potential μ_0^i has its own electromechanical coupling, such that Eq. (4) results in

$$\mu^i(x) = \mu_0^i + \hbar\omega\lambda^i x. \quad (11)$$

For simplicity, we neglect any gate-voltage dependence of the mechanical frequency of the CNT, i.e., $\omega^i = \omega \forall i$. This assumption is consistent with experimental observations (see Appendix D). Due to each electromechanical coupling, the displacement of the CNT is altered by the electronic transport through each of the multiple electrochemical potentials; thus, the equation of motion given [Eq. (7)] is extended to

$$\ddot{x} + \delta\dot{x} + \omega^2 \left[x + 2 \sum_i \lambda^i n^i(x) \right] + \epsilon x^3 = f_{\text{ex}} \cos(\omega_{\text{ex}} t), \quad (12)$$

where $n^i(x)$ is the general version of Eq. (8) as

$$n^i(x) = \frac{1}{2} + \frac{\Gamma_R^i}{\pi \Gamma_{\text{tot}}^i} \left[\arctan(\gamma_R^i(x)) + \frac{\Gamma_L^i}{\Gamma_R^i} \arctan(\gamma_L^i(x)) \right]. \quad (13)$$

We note that we allow for each electrochemical potential μ_0^i to have its own tunneling parameters, i.e., $\Gamma_{L/R}^i$, and thus $\gamma_v^i(x) = 2(\mu_v - \mu^i(x))/(\hbar\Gamma_{\text{tot}}^i)$. As in previous sections, in order to compute the nonequilibrium steady state, we simultaneously solve Eqs. (11)–(13).

Here, we directly study the average current given that it is experimentally accessible, and we later compare it with measurements. In Appendix E, a detailed analysis of the CNT displacement can be found. To account for all the contribu-

tions to transport arising from the different electrochemical potentials, Eq. (10) is modified as

$$I_{\text{avg}} = \frac{1}{T} \lim_{T \rightarrow \infty} \int_{\tau}^{\tau+T/\omega_0} dt \sum_i I^i(x(t)) \quad (14)$$

with the instantaneous generalized current [from Eq. (9)]

$$I^i(x) = \frac{e}{\pi} \frac{\Gamma_L^i \Gamma_R^i}{\Gamma_{\text{tot}}^i} \left[\arctan(\gamma_L^i(x)) - \arctan(\gamma_R^i(x)) \right]. \quad (15)$$

We compare the predictions of our generalized model with measurements of electron transport in a suspended CNT device. Fabrication, experimental details, and data analysis can be found in Appendix C. Varying the amplitude of the driving power P_{ex} , we explore different driving regimes of the electromechanical system.

Following a similar approach to that in Sec. IIC, we start by probing the device response at low driving power P_{ex} , which corresponds to a low driving force F_{ex} [47]. Figure 4(a-1) shows the simulated I_{avg} [from Eq. (14)] as a function of gate voltage V_g (which displaces the QD electrochemical potential $\{\mu_0^i\}_i$) and driving frequency ω_{ex} . Figure 4(a-2) shows the measured current. In both cases, we observe Coulomb peaks corresponding to electrochemical potentials μ_0^{N+1} , μ_0^N , and μ_0^{N-1} , and, as expected, the mechanical resonance exhibits softening at each of these peaks. We only showcase the I_{avg} due to sweeping the ω_{ex} from low to high values (as indicated in with red arrows); see Appendix F for the results of the opposite sweeping direction.

Next, we increase the driving power P_{ex} (and hence force F_{ex}) in Fig. 4(b-1) [(Fig. 4(b-2))]. Here, for both the simulations and experiments, the softening of the mechanical resonance frequency is more evident and an archlike structure begins to develop (highlighted with blue arrows). Each of the peaks in I_{avg} can be traced back to the Coulomb peaks corresponding to μ_0^{N+1} , μ_0^N , and μ_0^{N-1} . In between two peaks (marked with a black cross), an enhancement of I_{avg} is observed. We attribute this feature to the fact that higher amplitudes of the CNT motion are achieved with a higher driving $P_{\text{ex}}(F_{\text{ex}})$, which in turn causes a larger shift in the electrochemical potentials of the QD, allowing for both μ_0^{N+1} and μ_0^N to enter the transport window within one cycle of the CNT oscillation (see inset). In this way, the combination of these transport channels contributes to an increase of the net current.

Finally, we further increase the driving power (force) to the strong driving regime [Figs. 4(c-1) and 4(c-2)]. Here, we see the arch-shaped structure fully develop. As detailed in the previous section, the observed archlike response is due to the Duffing nonlinear contribution.

We conclude this section by emphasizing that our model faithfully reproduces the nonlinear features observed in the electronic current across all external driving strengths within a single microscopic framework. As a final comment, we note that the experimental data showcase sudden current drops not present in the simulated data [see Figs. 4(b) and 4(c)]. We expect our model to additionally reproduce these drops if we were to include noise (i.e., electronic) by replacing our classical equation of motion [Eq. (7)] with a Langevin-type equation [48].

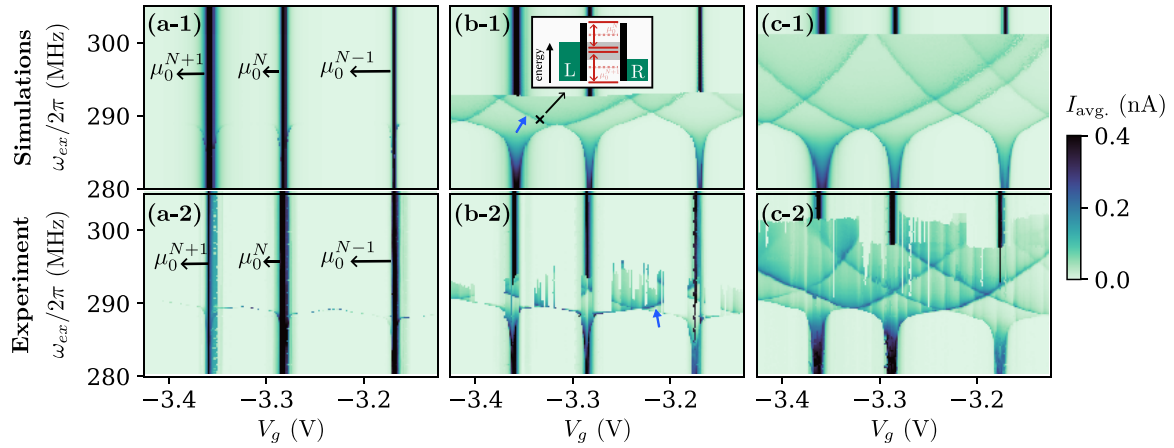


FIG. 4. Top panels (a-1)–(c-1) showcase the simulated average current as a function of the gate voltage V_g and the driving frequency ω_{ex} (from low to high values) following the model described in Sec. III. Left to right are for increasing external normalized driving forces: (a-1) $f_{ex}/\omega^2 = 0.1$, (b-1) $f_{ex}/\omega^2 = 2$, and (c-1) $f_{ex}/\omega^2 = 4$. Here, we consider three QD electrochemical potentials labeled as μ_0^{N+1} , μ_0^N , and μ_0^{N-1} . The electronic parameters used for these simulations are detailed in Table I, and the mechanical parameters are $\epsilon/\omega^2 = 9.38 \times 10^{-9}$ and $\delta/\omega = 7.7 \times 10^{-4}$. Inset in panel (b-1) shows the energy configuration corresponding to two electrochemical potentials entering the transport window in one cycle, which allows for an enhancement of the current (see main text). Bottom panels (a-2)–(c-2) are the measured average current as a function of the gate voltage V_g and the driving frequency ω_{ex} (from low to high values) with an applied bias of 0.25 mV, the tunneling and mechanical extracted parameters are detailed in Appendix D. Left to right describe an increase on driving power being (a-2) –76 dBm, (b-2) –66 dBm, and (c-2) –53 dBm. All the panels share the same color bar detailed in the right side of the figure.

IV. CONCLUSIONS

In this work, we studied the nonlinear response of an externally driven CNT mechanical resonator with a electrostatically defined QD. The system displays the interplay of two distinct sources of mechanical nonlinearities, namely, (1) coupling between the electronic transport and the motion, and (2) intrinsic mechanical contributions. We have presented a theoretical model that is able to capture these very different contributions by varying the external driving force.

To begin, we considered the case where the electronic transport is given by a single electrochemical potential in the QD. We found that, in the weak driving regime, the predominant nonlinearity is given by the electromechanical coupling, source (1). In such case, the resonance frequency presents the well-known softening dip given by the electromechanical coupling strength λ , consistent with previous works [16,18,21]. As the driving power is increased, we found that these dips persist although the mechanical nonlinearities, source (2), start to dominate. A broadening of the resonance frequency was observed and it presented a frequency sweep hysteresis. In the strong driving regime, where source (2) dominates, the displacement response of the CNT is of a Duffing type. Moreover, in the current through the device, we observed the development of archlike structures. This archlike appearances constitute the main feature of the presented model in the strong driving regime.

We then extended our model to account for multiple electrochemical potentials of the QD inside of the bias window. We finally compared the predictions with device measurements of the current through a CNT and observed that they successfully qualitatively reproduce the experimental results and capture features that had previously been overlooked.

This work provides an essential characterization of the carbon nanotube response across all driving strengths, paving the way for future exploration of nonlinear phenomena. Future

directions may include the inclusion of thermal and electronic noise to the model, as well as the study of synchronization events and the extension to the quantum regime.

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S.S.: Model development (equal); Simulations (lead); Visualization (lead); Data Analysis (lead); Writing—original draft (lead); Writing—review and editing (equal). K.A.: Experimental data (lead); Visualization (supporting); Writing—original draft (supporting); Discussions (equal). J.T.-B.: Discussions (equal); Writing—review and editing (equal). J.M.: Model development (equal); Simulations (supporting); Discussions (equal); Writing—review and editing (equal). F.V.: Fabrication (lead); Writing—review and editing (equal). F.F.: Discussions (equal); Writing—review and editing (equal). J.D.: Discussions (equal); Writing—review and editing (equal). J.M.R.P.: Discussions (equal). G.J.M.: Model development (equal); Discussions (equal). J.A.: Supervision (equal); Discussions (equal); Writing—review and editing (equal). N.A.: Supervision (equal); Discussions (equal). F.C.: Model development (equal); Simulations (supporting); Visualization (supporting); Data Analysis (supporting); Writing—review and editing (equal).

DATA AVAILABILITY

The codes used for the present work as well as the measured experimental dataset and analysis that support the findings of this study are available in Ref. [49].

APPENDIX A: DERIVATION OF THE CNT EQUATION OF MOTION

In this Appendix, we detail how to obtain Eq. (7). To do so, we use Hamilton's equations for the full Hamiltonian detailed in Eq. (1). This results in the following classical equation of motion:

$$m\ddot{z} + m\delta\dot{z} + m\omega^2 z + \frac{\hbar\omega\lambda}{z_{\text{ZPM}}} n(z(t)) + \frac{\beta}{z_{\text{ZPM}}} \left(\frac{z}{z_{\text{ZPM}}} \right)^3 = F_{\text{ex}} \cos(\omega_{\text{ex}} t). \quad (\text{A1})$$

Note that we have included a friction term $m\delta\dot{z}$ to account for the intrinsic sources of dissipation of the mechanical oscillator. Then, defining the normalized displacement with respect to the zero-point motion, $x = z/z_{\text{ZPM}}$, one arrives to Eq. (7).

APPENDIX B: NUMERICAL CALCULATION OF THE NONEQUILIBRIUM STEADY STATE

In order to obtain the nonequilibrium steady state of the semiclassical system, we simultaneously solve Eqs. (7) and (8). This was done by implementing the code available in Ref. [49] written in Julia (v1.12). We have made use of the “DifferentialEquations.jl” (v7.17.0) [50]. The solver used is “Verner's ‘Most Efficient’ 9/8 Runge-Kutta method” with initial time step 1×10^{-10} , absolute tolerance 1×10^{-5} , and relative tolerance 1×10^{-5} . To proceed, it is necessary to supply the code with initial conditions, namely, $x(0)$ and $\dot{x}(0)$. As we are dealing with nonlinear equations whose solutions depend sensitively on the direction of the external driving

frequency sweep ω_{ex} , different initial conditions must be specified to avoid convergence to the same steady-state solution. For simulations where ω_{ex} is swept from low to high values, we use as initial conditions the frequency response of a forced Duffing oscillator $A(\omega_{\text{ex}})$, obtained by setting $\lambda = 0$ in Eq. (7) [see also Eq. (D2)]. To produce the data, we have used the “oscillator.jl” code provided in Ref. [51]. We find that computing the response at the highest driving force is sufficient, and the resulting steady-state displacement can then be used as the initial condition for simulations at smaller forces. Therefore, the initial conditions is $x(0) = A(\omega_{\text{ex}})$ and $\dot{x}(0) = 0$. Conversely, for the reverse sweep of ω_{ex} (from high to low frequencies), we set the initial conditions to be $x(0) = 0$. The initial states used for the simulations discussed in Secs. II B and II C, together with the corresponding frequency responses at smaller driving forces, are shown in Figs. 5(a)–5(c).

Next, we integrated the full equations of motion [Eqs. (7) and (8)], obtaining the dynamics of the CNT for long times until the nonequilibrium steady state was reached, i.e., about $5000\pi/\omega$ for the parameters used. In Fig. 5(d), we plot an example of the dynamics for the parameters of Fig. 2. Here, we choose $\omega_{\text{ex}}/\omega = 1$ and $\mu_0 = 0$ meV. As the CNT is externally driven, we expect the nonequilibrium steady state to be of the form $x(t \rightarrow \infty) = x^{\text{NESS}} \cos(\omega_{\text{ex}} t)$. The amplitude x^{NESS} is then obtained from the amplitude of long-time dynamics, as shown in Fig. 5(e).

APPENDIX C: DEVICE FABRICATION AND EXPERIMENTAL DETAILS

The device is a nanoelectromechanical resonator formed by suspending a carbon nanotube on metal electrodes [Fig. 6(a)]. A bias voltage $V_{\text{SD}} = \Delta\mu/e$ is applied from source to drain electrodes, driving a current I_{DC} through the nanotube. The carbon nanotube is suspended above five gate electrodes with voltages $V_{\text{G1-G5}}$. The mechanical oscillations of the nanotube are excited by a radio frequency (rf) drive, with frequency f_{ex} and driving power P_{ex} , connected to one of the gates. The gates are further used to define a QD along the carbon nanotube. All experiments in this work are carried out in a dilution refrigerator with a base temperature of 60 mK.

The suspended carbon nanotube device is the same as the one presented in Ref. [16] but measured in a different cooldown. The device is fabricated on a high resistance Si/SiO₂ substrate by patterning Au/Cr electrodes with electron beam lithography. Carbon nanotubes are grown by chemical vapor deposition on a separate quartz substrate using nanoparticles of Al₂O₃, Fe(NO₃), and MoO₂(acac)₂ as catalyst and mechanically transferred to the Si chip.

APPENDIX D: CHARACTERIZATION OF THE DEVICE

Now, we detail how we extracted the tunneling and coupling parameters from the experimental data to later employ in the model and numerical calculations.

We first extract the lever arm $\alpha = eC_{\text{G}}/C$, where C_{G} is the capacitance between the gate and the QD, and $C = C_{\text{G}} + C_{\text{S}} + C_{\text{D}}$ is the total capacitance between the gate, source, and drain. Similarly to Ref. [16], we calculate the two slopes $\alpha_1 = -eC_{\text{G}}/(C - C_{\text{S}})$ and $\alpha_2 = eC_{\text{G}}/C_{\text{S}}$ of the Coulomb

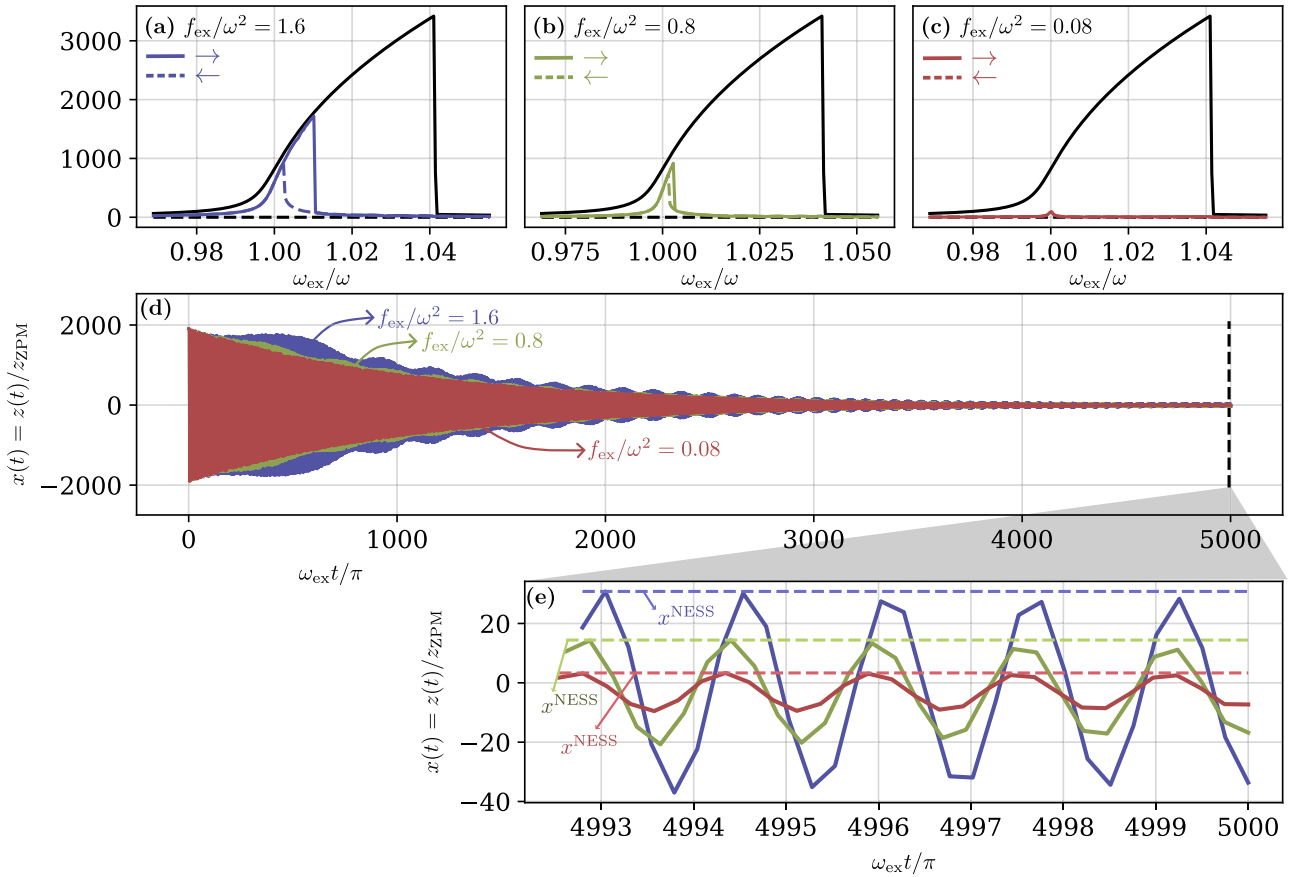


FIG. 5. The initial states (black) together with the corresponding frequency responses at smaller driving forces as (a) $f_{\text{ex}}/\omega^2 = 1.6$ (purple), (b) $f_{\text{ex}}/\omega^2 = 0.8$ (green), and (c) $f_{\text{ex}}/\omega^2 = 0.08$ (red). Solid lines indicate the external driving frequency ω_{ex} sweep from low to high values, and the dashed lines indicate the opposite direction. (d) Time dynamics for the displacement of the CNT with respect to its zero-point motion z_{ZPM} for three external driving forces: $f_{\text{ex}}/\omega^2 = 1.6$ (purple), $f_{\text{ex}}/\omega^2 = 0.8$ (green), and $f_{\text{ex}}/\omega^2 = 0.08$ (red). In these panels, the driving frequency ω_{ex} was set to 1 Hz and the QD energy to $\mu_0 = \text{meV}$. (e) Close-up to large timescale highlighted with a black dashed line in panel (a). With horizontal dashed lines, we characterize the nonequilibrium steady-state displacement of the CNT for each driving force. Parameters: $\delta/\omega = 9.4e-9$, $\epsilon/\omega^2 = 8e-4$, $\lambda = 3$, $\mu_L = -\mu_R = 0.1 \text{ meV}$, and $\Gamma_L/2\pi = \Gamma_R/2\pi = 30 \text{ GHz}$. Additionally, for solid line initial condition in panel (a), $f_{\text{ex}}/\omega^2 = 4$.

diamonds (without an external driving) as shown in Fig. 6(b). The lever arm α is determined as [52]

$$\alpha = \frac{\alpha_1 \alpha_2}{\alpha_1 - \alpha_2} = 0.083 \text{ eV/V}. \quad (\text{D1})$$

Next, we extract the tunneling parameters for the QD electrochemical potentials $\mu_0^{N+2} - \mu_0^{N-2}$. To estimate the tunneling parameters for the bias voltage $V_{\text{SD}} = 0.25 \text{ mV}$ used in Sec. III, we average the value by fitting Coulomb peaks to the nearby values of $V_{\text{SD}} = 0.2 \text{ mV}$ and $V_{\text{SD}} = 0.275 \text{ mV}$. The measured Coulomb peaks are presented in Fig. 6(c) (with external driving $P_{\text{ex}} = 0$) from which the tunneling rates are obtained by fitting each peak to Eq. (9) and setting $\lambda = 0$. In Fig. 6(d), we show the results of the fits.

To compute the electromechanical coupling strength $g_m = \omega\lambda$, we turn on the external drive with power $P_{\text{ex}} = -45 \text{ dBm}$ and follow the same procedure as in Ref. [16]. Due to the multiple electrochemical potentials of the QD participating in the transport, we determine individually each of the electromechanical coupling strengths g_m^i , corresponding to the i th electrochemical potential. In Fig. 7(a), we showcase the frequency response of the CNT as a function of the gate

voltage V_g and external driving frequency ω_{ex} . From here, we can extract the softening of the mechanical resonance of the peaks $\mu_0^{N+1} - \mu_0^{N-1}$. We find the values of $\{g_m^i\}_{i=N+1, N, N-1}$ by qualitatively matching the experimental data as shown in Fig. 7(b). The extracted parameters can be found in Fig. 7(c). The parameters for peaks corresponding to μ_0^{N+2} and μ_0^{N-2} were estimated based on experimentally realistic values. Note that the resonance mechanical frequency of the carbon nanotube ω for the different electrochemical levels ranges from $2\pi \times 288 \text{ MHz}$ to $2\pi \times 289.7 \text{ MHz}$. Given this narrow variation, we adopted the intermediate value $\omega = 2\pi \times 289 \text{ MHz}$ for our theoretical simulations in Sec. III.

The final step is to characterize the Duffing parameter ϵ and damping δ of the CNT as defined in Eq. (7). In principle, these parameters should be obtained by fitting the displacement of the CNT. However, in the experiment, we only have access to the current I_{DC} through the nanotube. We therefore estimate these parameters so as to be able to reproduce the current intensity and broadening observed in the experiment at a fixed gate voltage V_g . Figure 8(a) shows the current I_{DC} for driving power $P_{\text{ex}} = -53 \text{ dBm}$ and $V_g = -3.34 \text{ V}$. We see that the current I_{DC} presents three peaks, corresponding to the

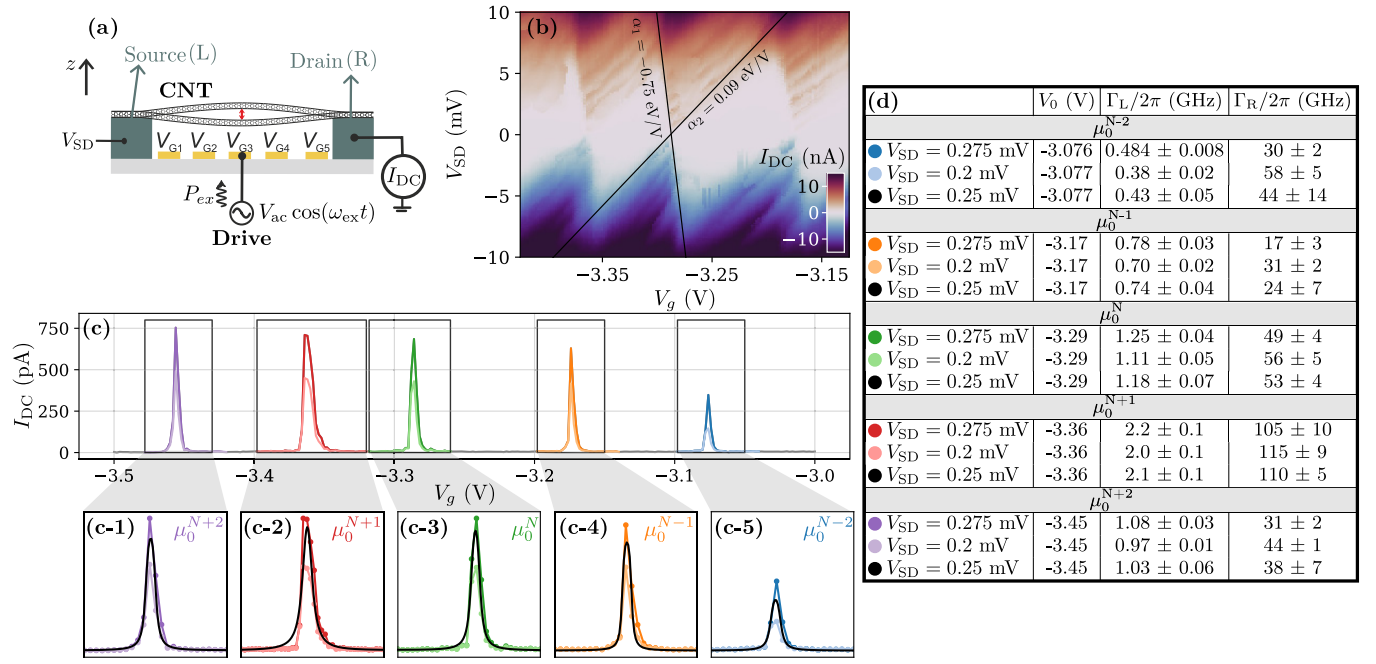


FIG. 6. (a) Schematic of the experimental device. A carbon nanotube is suspended between the source and drain metal electrodes. Five gate electrodes with voltages V_{G1-G5} are used to define a quantum dot. A bias voltage $V_{SD} = \Delta\mu/e$ drives a current I_{DC} through the carbon nanotube. An rf drive $V_{ac} \cos(\omega_{ex}t)$ with power P_{ex} excites the mechanical motion of the carbon nanotube. (b) Measured I_{DC} as a function of V_{SD} and $V_g = V_{G1}$ showing Coulomb diamonds. The slopes α_1 and α_2 of the Coulomb diamonds, highlighted in black, are used to estimate the lever arm α . (c) Coulomb peaks for the range of $V_g = V_{G1}$ under consideration in the main text. The darker color trace corresponds to $V_{SD} = 0.275$ mV, and the lighter color trace corresponds to $V_{SD} = 0.2$ mV. The bottom panel shows a close-up of the Coulomb peaks. The solid black lines fits evaluated using Eq. (9) (and setting $\lambda = 0$) with parameters exhibited in panel (d).

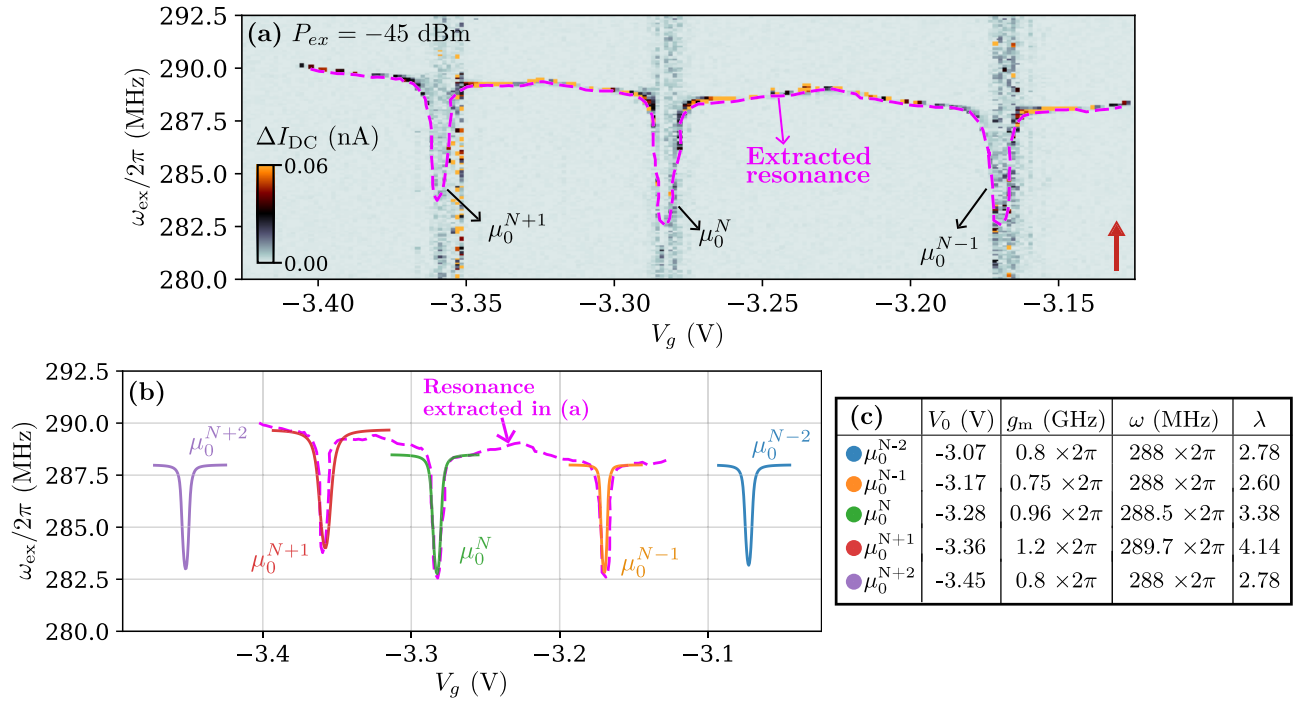


FIG. 7. (a) Differential current response to the lowest driving power, i.e., -45 dBm, as a function of gate voltage V_g and driving frequency ω_{ex} swept from low to high values as indicated with a red arrow. The extracted resonance for the $\mu_0^{N+1} - \mu_0^{N-1}$ QD electrochemical potentials is highlighted with a dashed magenta line. (b) Softening of the mechanical resonance for different Coulomb peaks extracted from panel (a) (shown in dashed magenta). The solid lines show the calculated softening of the mechanical resonance using parameters in panel (c) following Ref. [16]. (c) Parameters characterizing the coupling of mechanical motion to the electron transport obtained from panel (a). The parameters for peaks corresponding to μ_0^{N-2} and μ_0^{N+2} are estimated based on experimentally realistic values.

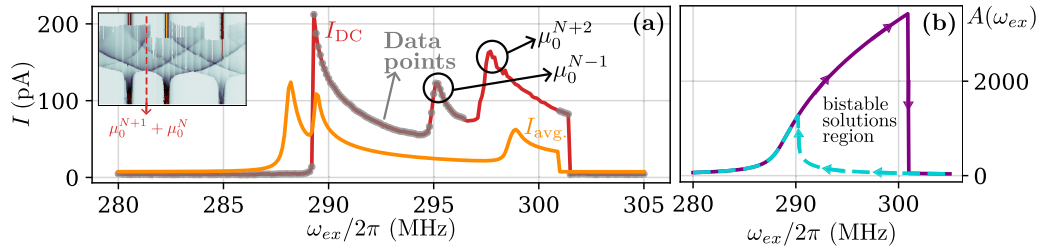


FIG. 8. (a) I_{DC} as a function of driving frequency ω_{ex} . The red trace is the measured mechanical resonance for driving power $P_{ex} = -53$ dBm and gate voltage $V_{G1} = -3.34$ V, when the Duffing nonlinearity is prominent. The yellow trace is the calculated I_{avg} , calculated using the guess values ($\epsilon/\omega^2 = 9.38 \times 10^{-9}$ and $\delta/\omega = 7.7 \times 10^{-4}$) and normalized force $f_{ex}/\omega^2 = 4$. (b) Displacement of the CNT with the simplification made in Eq. (D2) used to compute I_{avg} of panel (a).

QD electrochemical potentials $\mu_0^{N+1} + \mu_0^N$, μ_0^{N-1} , and μ_0^{N+2} . The various parameters characterizing the electron transport through the QD and the mechanical motion of the CNT are intricately linked to each other. Therefore, finding each value of these parameters for all the combinations of the others becomes an onerous challenge. Thus, we make a few simplifications to estimate the resonator parameters:

(1) In the experiment, the frequency of the mechanical resonance of the nanotube changes as a function of the gate voltage V_g [see Fig. 7(a)]. The first simplification that we consider is that the mechanical resonance frequency ω of the CNT is independent of V_g , such that for all peaks $\omega/2\pi = 289$ MHz. Experimentally, this can be seen in Fig. 4 as the overall linear trend of the frequency to decrease as V_g increases. Indeed, this is why in the simulations after each dip the frequency goes back up to the same value as before, while in the experiment it goes to a value slightly smaller. The dependence of the frequency on V_g has been extensively studied previously elsewhere (see, e.g., Refs. [21,22]). Here, we choose to exclude it from our model to simplify the number of free parameters, since except for this overall trend, it does not affect the shape of the frequency dips or the broadening effects that are the main focus of this work. Adding this effect to our model could be done straightforwardly.

(2) Since Eqs. (12)–(15) do not allow for a simple analytical expression for the current in the steady state, we consider that in the nonequilibrium steady state, the amplitude of the CNT follows the response of a Duffing oscillator, which is

$$z(t) = A(\omega_{ex}) \cos(\omega_{ex}t) \quad \text{with} \quad A(\omega_{ex}) = \frac{f_{ex}}{\sqrt{(\omega_{ex}^2 - \omega^2 - \frac{3}{4}\epsilon A^2(\omega_{ex}))^2 + (\delta\omega_{ex})^2}}. \quad (\text{D2})$$

Then, given ϵ and δ , the nonequilibrium steady-state current is given by replacing this value of z in Eqs. (11) and (15).

Under these simplifications, for every value of ϵ and δ , we produce an approximate current response and compare with the experimental measurements. In Fig. 8(a), in orange, we plot the result for the best set of values, $\epsilon/\omega^2 = 9.38 \times 10^{-9}$ and $\delta/\omega = 7.7 \times 10^{-4}$. The corresponding displacement of the CNT using Eq. (D2) is shown in Fig. 8(b). We find that with these simplifications, the model is qualitatively able to capture the role of different nonlinearities in the experimental data. This allows us to understand a hybrid nanoelectromechanical system with a simple theoretical model.

In Table I, we summarized all the parameters used in order to compute Figs. 4(a-1)–4(c-3).

It is important to note that in our experimental dataset, we observe both softening and hardening of the mechanical resonance. This is not captured in the model because the Duffing parameters are fixed to one of these cases. We attribute this change of value and sign of the nonlinearity to different gate voltage configurations and driving power, as this phenomenon has been previously observed and characterized in both CNT and other nanomechanical systems [18–20,53].

APPENDIX E: CNT DISPLACEMENT FOR THE ESTIMATED DEVICE PARAMETERS

In Fig. 9, we plot the CNT displacement for the parameters estimated from the experimental measurements shown in Fig. 4 as a function of the gate voltage V_g and the driving frequency ω_{ex} , and for increasing external driving force. The interpretation of the observed behavior is analogous to what was discussed in Sec. II B for the single electrochemical potential case. Here, we moreover incorporate the effect of having multiple QD electrochemical potentials within the bias window, including the effect of the backaction on the CNT displacement from each possible transition, as was covered in detail in Sec. III.

APPENDIX F: BISTABLE RESPONSE AND FREQUENCY SWEEPING HYSTERESIS

In Fig. 4, we showed the experimentally measured current and the model predictions when the frequency ω_{ex} of the external drive is swept from low to high frequency. In Fig. 10, we now plot instead the calculated (top panels) and measured

TABLE I. Summary of parameters extracted from the device used to compute the simulations in Figs. 4(a-1)–4(c-1). Note that there is no need to estimate the value of z_{ZPM} and the mass m of the CNT in this model.

	V_0 (V)	$\Gamma_L/2\pi$ (GHz)	$\Gamma_R/2\pi$ (GHz)	λ	ω (MHz)
μ_0^{N-2}	−3.07135	0.430264	44.2383	2.78	$289 \times 2\pi$
μ_0^{N-1}	−3.16863	0.737844	24.0938	2.60	$289 \times 2\pi$
μ_0^N	−3.2814	1.17658	52.808	3.38	$289 \times 2\pi$
μ_0^{N+1}	−3.35679	2.13673	110.354	4.14	$289 \times 2\pi$
μ_0^{N+2}	−3.45068	1.02658	37.7511	2.78	$289 \times 2\pi$

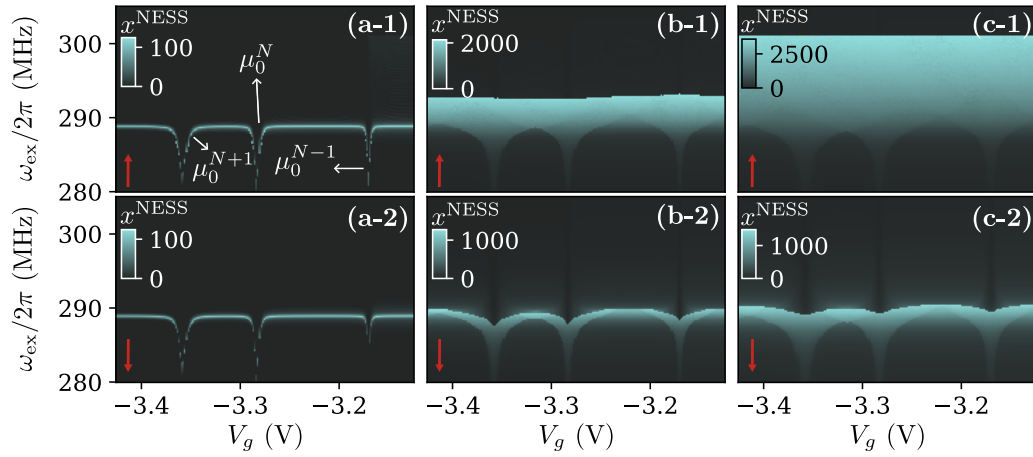


FIG. 9. Displacement of the CNT with respect to its zero-point motion $x^{\text{NESS}} = z^{\text{NESS}}/z_{\text{ZPM}}$ in the nonequilibrium steady state as a function of both the external driving frequency ω_{ex} and gate voltage for increasing normalized driving forces as (a-1)/(a-2) $f_{\text{ex}}/\omega^2 = 0.1$, (b-1)/(b-2) $f_{\text{ex}}/\omega^2 = 2$, and (c-1)/(c-2) $f_{\text{ex}}/\omega^2 = 4$. Top panels correspond to the external driving frequency ω_{ex} driving from low to high powers (per indicated with a red arrow) and bottom panels correspond to the ω_{ex} swept in the opposite direction.

(bottom panels) I_{avg} response when the external driving frequency ω_{ex} is swept from high to low frequency. Due to the strong nonlinearity of the system, a hysteresis in the response is observed. The dependence on sweeping direction of the

measured I_{avg} is weaker for the lowest driving force (power) in Fig. 10(a-1) [Fig. 10(a-2)] and becomes noticeably significant as the driving force (power) increases in Figs. 10(b-1) and 10(c-1) [Figs. 10(b-2) and 10(c-2)].

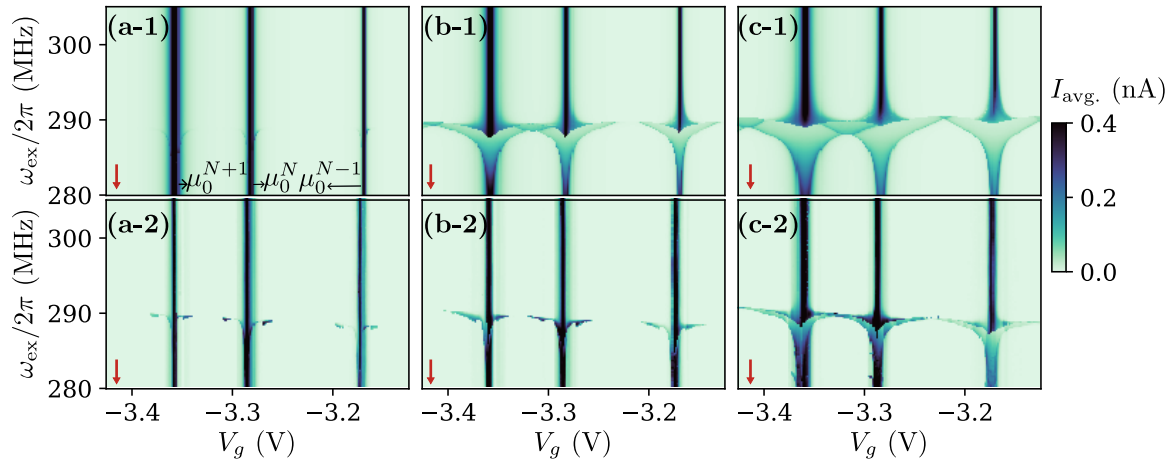


FIG. 10. Similar to Fig. 4, however, the external frequency ω_{ex} is swept from high to low values. The top panels (a-1)–(c-1) show the calculated I_{avg} for external normalized driving forces $f_{\text{ex}}/\omega^2 = 0.1$, $f_{\text{ex}}/\omega^2 = 2$, and $f_{\text{ex}}/\omega^2 = 4$, respectively. While the bottom panels (a-2)–(c-2) showcase the measured current I_{DC} for different driving powers $P_{\text{ex}} = -76$ dBm, $P_{\text{ex}} = -66$ dBm, and $P_{\text{ex}} = -53$ dBm, respectively.

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