Real-space tailoring of the electron–phonon coupling in ultraclean nanotube mechanical resonators

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The coupling between electrons and phonons is at the heart of many fundamental phenomena in nature. Despite tremendous advances in controlling electrons or phonons in engineered nanostructures, control over their coupling is still widely lacking. Here we demonstrate the ability to fully tailor electron–phonon interactions using a new class of suspended carbon nanotube devices, in which we can form highly tunable single and double quantum dots at arbitrary locations along a nanotube mechanical resonator. We find that electron–phonon coupling can be turned on and off by controlling the position of a quantum dot along the resonator. Using double quantum dots we structure the interactions in real space to couple specific electronic and phononic modes. This tailored coupling allows measurement of the phonons’ spatial parity and imaging of their mode shapes. Finally, we demonstrate coupling between phonons and internal electrons in an isolated system, decoupled from the random environment of the electronic leads, a crucial step towards fully engineered quantum-coherent electron–phonon systems.

Some of the most well-known phenomena in molecular and solid-state physics result from the coupling between electrons and phonons. The resistivity of metals, ferroelectricity, Peierls and Jahn–Teller instabilities and BCS superconductivity are different facets of this coupling. In solids, electron–phonon coupling is dictated by the lattice structure and the ensuing electronic bands, leaving little room for tunability. Over the past decades, tremendous advances have been made in the ability to engineer materials on the nanoscale. On the electronic side, artificial atoms—quantum dots—were created, providing extensive control over their electronic spectrum, which allowed the exploration of a wide variety of phenomena inaccessible in bulk solids. On the mechanical side, a growing variety of engineered systems enabled the study of mechanical phenomena on the nanoscale and brought experiments closer towards controlling the quantum state of mechanical resonators, as well as their coupling to single spins, qubits and photons. These remarkable advances contrast with the still-limited control over the coupling between the electronic and mechanical subsystems, whose tailoring would provide a remarkable toolbox for nano-electro-mechanical systems in the classical and quantum regimes.

Carbon nanotubes constitute a particularly promising system for tailoring the electron–phonon coupling. Their pristine lattice recently enabled the realization of extremely clean electronic systems, albeit still limited in length and complexity. Moreover, their one-dimensional nature, light mass and large stiffness enabled the creation of tunable mechanical resonators with high Q-factors. Recent pioneering works coupled a carbon nanotube resonator to a single quantum dot, demonstrating that the mechanical frequency can be strongly affected by a single carrier and that the correlated motion of electrons and vibrations can lead to mechanical frequency softening.

In this work, we explore a new generation of suspended carbon nanotube devices with wide-ranging local control, allowing the formation and manipulation of single and double quantum dots embedded into the mechanical oscillator. Using this unprecedented level of control we engineer local field gradients that produce controllable coupling between the electrons and the acoustic transverse phonons. The advantage of this coupling over the intrinsic strain-induced electron–phonon coupling is that it can be tailored at will. Our measurements are done in the classical limit (large vibration amplitudes) but because the interaction with the external fields is linear in the carbon nanotube displacement, these measurements probe the same electron–phonon coupling matrix element that would be relevant in the quantum limit. We study the dynamics of this coupling, and its spatial dependence, as well as demonstrate mode-selective coupling in an isolated system.

The device (Fig. 1a) is created using the nano-assembly technology we recently developed. It consists of a small-bandgap carbon nanotube suspended between two metallic contacts above five electrically independent gates. Above the metallic contacts the carbon nanotube is hole-doped due to the contacts’ workfunction. The suspended part, on the other hand, can be locally doped with either electrons or holes by applying independent d.c. voltages to the gates, \( V_{g1} \) to \( V_{g5} \). A negative voltage on all gates dopes the entire carbon nanotubes with holes, effectively creating a continuous ‘wire’ whose conductance is only weakly gate-dependent. However, when a positive voltage is applied to one of the gates while keeping negative voltages on the others, the nanotube’s segment above this gate is doped by electrons, forming a pair of p–n junctions that confine a quantum dot above this gate (Fig. 1b).

As we will show, not only does this dot act as a detector of the local mechanical motion through its charge sensitivity, but more importantly, it provides controlled local coupling between the electronic and mechanical systems, which forms the fundamental building block for this work.

The mechanical vibrations are measured through a standard mixing technique. A radiofrequency signal with frequency \( f \)
is applied to an off-centre gate (gate 4) and a weaker ‘probe’ signal with frequency $f + \delta f$ is applied to the source electrode (Fig. 1b). The former actuates the mechanical motion and mixes down with the latter through the dependence of the current on the source–drain and gate voltages, $\delta^2 I/\delta V_{SD} \delta V_g$, to produce a low-frequency ($\delta f$) current signal measured at the drain. When $f$ is swept through a mechanical resonance, the carbon nanotube vibration is enhanced, producing a sharp peak in the out-of-phase quadrature of the mixing signal, $M_y$, as well as in the derivative of its in-phase quadrature with respect to $f$, $dM_y / df$ (Fig. 1c, more details in Supplementary Information 1). Here, we use these peaks interchangeably to trace out the mechanical resonance.

Figure 1d,e shows the measured gate dependence of the first two mechanical modes of the carbon nanotube resonator. The motion is detected by a quantum dot formed on the resonator at a position of large movement (above gate 3 for the first mode and above gate 4 for the second mode, see illustrations). The bottom panels show the measured $M_y$ (colour map) as a function of the voltage on the gate beneath the dot and of the drive frequency, $f$; the top panels show the simultaneously measured conductance. For both modes the mixing signal is visible whenever the dot is conducting and, being proportional to the derivative of the conductance with respect to gate voltage, it is negative on one side of the Coulomb peak (blue), positive on the other (red), and zero at the peak (white). As in previous experiments, we observe that for both modes, the resonance frequencies increase with gate voltage owing to the tensioning of the resonator, and that the coupling between the electronic and mechanical degrees of freedom causes a sharp softening dip of the resonance frequency concomitant with the Coulomb peak. This dynamical coupling will be used here to control the interactions between the mechanical and electronic degrees of freedom.

The first step in creating a tailored interaction between electrons and phonons is to study the underlying dynamics of their coupling, so far hardly explored experimentally owing to the limited control over tunnelling barriers. Fundamentally, this coupling results from correlated mechanical and electronic motions: owing to carbon nanotube vibrations, electrons are pumped between the leads and the quantum dot and their attraction to a biased gate causes a softening of the mechanical restoring force. This process involves an interesting competition between the vibrational frequency and the electronic tunnelling rates, which we study here using a tunable-barrier quantum dot, formed in the resonator over the three central gates and populated with holes (Fig. 2a). The quantum dot can be tuned across the entire range from a closed quantum dot to the open Fabry–Perot-like regime, with individual control over the left and right tunnelling rates, $I_{\Gamma_1}$ and $I_{\Gamma_2}$, by the side gates (1 and 5). We calibrate these rates using transport measurements (Fig. 2b, details in Supplementary Information 2) and measure their independent effect on the mechanical softening. Starting with the symmetric case, $I_{\Gamma_1} \approx I_{\Gamma_2}$, we observe that the softening of the first mechanical mode, $\Delta f_1$, drops with progressive pinching-off of the barriers (Fig. 2c I–III). Plotting the extracted $\Delta f_1$ versus the total tunnelling rate $I_1 + I_2$ (Fig. 2d), we observe that the drop commences when the total tunnelling rate becomes comparable to the vibrational frequency, $2\pi f_1$. This drop reflects the inability of electrons to follow the mechanical motion and is reproduced by a theoretical calculation (dashed line, details in Supplementary Information 5). Interestingly, even when only one barrier is open ($I_{\Gamma_1} \gg 2\pi f_1 \gg I_{\Gamma_2}$) the softening assumes the maximal value (Fig. 2c IV). This demonstrates that contrary to the device conductivity, which is determined by the two barriers adding in series, the softening is controlled by the two rates added in parallel, reflecting that the relevant electrons can enter from either lead. Measurements at very large tunnelling rates (Supplementary Information 3) show...
Figure 2 | Dependence of dynamical electron–phonon coupling on the electron tunnelling rate. a, Schematics: a quantum dot of holes is created above gates 2–4. Its left and right barrier tunnelling rates, \( \Gamma_L \) and \( \Gamma_R \), are controlled by \( V_{g1} \) and \( V_{g5} \). b, Measurement of the peak conductance at the Coulomb blockade transition from 5 to 6 holes, \( G_{\text{peak}} \), as a function of \( V_{g1} \) and \( V_{g5} \), from which \( \Gamma_L \) and \( \Gamma_R \) are independently extracted (see Supplementary Information 2 for details). c, Mixing signal of the first mode, \( dM_1/df \), as a function of \( V_{g5} \) and \( f \) for symmetric tunnelling rates, \( \Gamma_1 \approx \Gamma_K \) (subpanels I–III), and for asymmetric rates, \( \Gamma_1 \ll \Gamma_K \) (subpanel IV). d, Extracted softening, \( \Delta f_i \), as a function of the total tunnelling rate, \( f^1 = \Gamma_1 + \Gamma_K \). The dashed line is a fit to the theory in Supplementary Information 6. The dotted vertical line marks the angular frequency of the mechanical mode, \( 2\pi f_1 \). Inset: the inverse mechanical Q-factor, deduced from the frequency width of the resonance, plotted as a function of \( V_{g3} \), across a Coulomb peak. Dashed line: theory fit (Supplementary Eq. 19). The difference between the on- and off-peak dissipation gives the electronic contribution, \( Q_{\text{elec}}^1 \). Main panel: extracted \( Q_{\text{elec}}^1 \) as a function of \( \Gamma_1 + \Gamma_K \) for \( \Gamma_1 \approx \Gamma_K \). Dashed line: theory fit (Supplementary Eq. 20). Error bars in d,e indicate 1 s.d. in \( \Delta f_1 \) and in \( Q_{\text{elec}}^1 \), respectively.

A similar reduction of \( \Delta f_1 \), this time due to gradual disappearance of the Coulomb blockade phenomenon\(^\text{23}\). In between these two drops, there is a wide range of tunnelling rates for which the softening remains practically constant (Fig. 2d and Supplementary Fig. 4). In this regime electrons enter sufficiently fast to establish electrostatic equilibrium at all times but not so fast to broaden the Coulomb blockade peaks beyond their thermal broadening.

The dependence on the total tunnelling rate is also remarkable for the dissipation of the phonon modes. As shown in the main panel of Fig. 2e, the dissipation (inverse Q-factor) is non-monotonic as a function of the electronic tunnelling rate, peaking when this rate is comparable to the vibration frequency. This might come as a surprise because, as in previous experiments\(^\text{18–21}\), we find that the dissipation decreases monotonically as the gate voltage is detuned away from the Coulomb blockade peak, which is also associated with reduced electron tunnelling. Nevertheless, the peak as a function of electron tunnelling is quantitatively described by our theory (dashed line, theory Supplementary Information 5) and can be rationalized as a close analogue of the Debye peak familiar from dielectric relaxation or viscoelasticity. Usually, the Debye peak is observed as a function of an indirect parameter (temperature) that changes the rates in the system. Here, in contrast, we observe it by directly controlling the physical parameter—the tunnelling rates of the electrons. The full control that we achieve over the relative dynamics of the electrons and vibrations will be used below to study previously unattainable regimes of their coupling.

A key feature that allows us to tailor the coupling between electrons and phonons is the control over the real-space confinement of the electrons. With five gates we can localize a quantum dot at five different locations along the tube, and explore how its position affects the coupling. To eliminate spurious position-dependence effects, we ensure that all of the parameters that are relevant for softening are similar for dots formed at the different locations. Specifically, all dots have the same number of electrons, similar charging energies and similar gate couplings (Supplementary Information 4), and their tunnelling rates are chosen well within the range where they do not affect the softening, as explained above. Interestingly, however, when measuring the softening of the first phonon mode with dots at the five locations (Fig. 3a–e, illustration in each panel) we observe that it depends strongly on position: it is weak near the contacts and increases continuously until reaching a maximum at the centre of the resonator. Figure 3f–j shows similar measurements for the second phonon mode. Again we observe strong position dependence; however, in contrast to the first mode, the softening is practically zero at the centre, and has its maxima above gates 2 and 4. Plotting the extracted softening for both modes, \( \Delta f_2 \) and \( \Delta f_4 \), as a function of the spatial position of the quantum dot (Fig. 3k,l), we find that they nicely follow the spatial displacement profile squared of the corresponding phonon modes (plotted as lines). The coupling thus provides a direct imaging of the shapes of the phononic modes in real space.

The observed spatial dependence can be understood by calculating the local forces generated on the carbon nanotube resonator due to single-electron charging of a localized quantum dot. In Supplementary Information 7 we show that this force can be described by an effective ‘electronic spring’ with a negative spring constant, ‘attached’ at the position of the dot (illustrated in Fig. 3k). The spring constant does not depend on the position of the dot along the tube, but the shift in the frequency of the combined system does: if the spring is connected at a node of the phononic mode it has no effect on its dynamics, whereas if it is connected at a location of large vibrational amplitude, it has a strong effect. Indeed, perturbation theory shows (Supplementary Information 7)
Figure 3 | Spatial dependence of the electron–phonon coupling, and direct imaging of the phonon modes. a–e, Mixing signals of the first phonon mode, $dM_1/df$ (colour map), measured with quantum dots formed above each of the five gates (see illustrations). Vertical bars are 0.5 MHz. The softening shows a clear dependence on the quantum-dot position. f–j, Similar measurements for the second mode, yielding a different position dependence. k, l, The softening of the first and second modes, $\Delta f_1$ and $\Delta f_2$, extracted from a–j and plotted as a function of the positions of the quantum dots, taken from the lithographic positions of the gates. Lines: calculated amplitude squared of the corresponding phonon mode versus position in the beam (solid) and string (dashed) limits (Supplementary Information 4). Inset to k, effective mechanical model: the force exerted on the nanotube by electrons flowing through a localized dot is equivalent to a spring with a negative spring constant attached at the dot’s position. Error bars in k, l: x axis bars indicate uncertainty in the location of the dot due to fabrication misalignment, and y axis bars indicate 1 s.d. in $\Delta f_1$.

Figure 4 | Tailored selective coupling between phononic and electronic modes in a double quantum dot. a, Conductance, G (colour map), of a double dot defined above gates 2 and 4 measured as a function of $V_{g2}$ and $V_{g4}$. The right, centre and left barriers are controlled by gates 1, 3 and 5, respectively. The label $G$ ($e^2/h$) represents the number of electrons in each dot. b, Zoom-in on the (3, 4) to (4, 3) transition. The common mode and detuning gating directions are labelled by V and $\epsilon$. c, First-mode mixing, $M_1$ (colour map), measured over the same voltage window as in b, d. The same for the second mode. In both cases we subtract the electronic mixing signal measured away from the mechanical resonance and integrate over a small frequency window to project out the frequency shifts due to softening (Supplementary Information 1). e, The mechanical motion of the different modes leads to different modes of effective gating; for $V_{g2} = V_{g4} = V$, the first-mode vibrations lead to common-mode gating (blue arrow) and the second mode leads to detuning gating (green arrow). f, g, Numerical derivatives of the conductance in b along the V and $\epsilon$ directions, respectively, showing good agreement with the measured mechanical mixing (c, d).
that the frequency shift is proportional to the amplitude squared of the bare phonon mode at the location of the local spring, in agreement with our observation. Beyond providing direct imaging of the phonon modes, the above measurements demonstrate that by moving the quantum dot in real space it is possible to turn on and off its coupling to selected phonon modes, thereby creating controllable coupling between these degrees of freedom.

We now take this coupled system to a new level of control, by embedding for the first time a double quantum dot in a mechanical carbon nanotube resonator. The added degrees of freedom allow us to tailor selective couplings between specific electronic and phononic modes, without the need to move the confinement potential. The observed physics has interesting connections to the coupling of electrons in double quantum dots and photons reported recently. The double dot is formed above gates 2 and 4, which also act as the corresponding plunger gates, and its left, right and centre tunnel barriers are controlled by the remaining three gates. The measured double-dot conductance as a function of the left and right plunger gate voltages (Fig. 4a) shows an extremely clean charge stability diagram, down to the single-electron limit. For the experiment we zoom onto a symmetric charge transition vertex (Fig. 4b) and study the two complementary facets of the electron–phonon coupling. We start by exploring the effects of the phonons on the electrons, through which we demonstrate the mode selectivity of this coupling. We then demonstrate the complementary effect of internal double-dot electronic modes on the phonons.

The coupling effects on the electrons are imprinted in the magnitude and sign of the mixing signal, as opposed to the frequency shift discussed so far. The former is isolated in Fig. 4c which plots the mixing signal $M_j$, measured for the first mechanical mode over the same voltage range as in Fig. 4b, but with the frequency shifts integrated out (see caption). A similar measurement for the second mechanical mode is shown in Fig. 4d. Curiously, we see that the patterns of negative and positive mechanical mixing signal (blue/red) are substantially different for these two modes. This difference reveals the distinct way that different phononic modes act on the electrons (Fig. 4e): in the first mode, the two dots are moving in phase, together getting closer and further away from the plunger gates. In the second mode, the dots move out-of-phase, with one approaching and the other receding from the gates. This different mechanical motion translates into different electrical gating: for equal d.c. voltages on the plunger gates, the first mode gates the double dot along the common-mode voltage direction (vector $V$ in Fig. 4b) whereas the second mode gates it along the detuning direction (vector $\varepsilon$ in Fig. 4b). Each phonon mode can thus be mapped onto an effective ‘gating vector’ in voltage space, and correspondingly, its mixing signal should be the derivative of the conductance along the direction of this vector. By taking the numerical derivative of the conductance in Fig. 4b along the $V$ and $\varepsilon$ directions (Fig. 4f), we clearly observe excellent agreement with the measured mixing signals of the first and second modes, respectively. The above measurement demonstrates two important aspects: first, it shows that it is possible to use the electrons to directly probe the real-space parity of the phonons. Furthermore, it demonstrates that each phonon mode has a characteristic action on the electrons, captured by its ‘gating vector’, providing a powerful tool for tailoring selective coupling between these degrees of freedom.

So far we have studied the different aspects of tailored coupling; however, this coupling was to random electrons tunnelling in and out from the leads. An even more interesting case would be to determine whether it is possible to couple the phonons to internal electrons in an isolated system, thereby realizing a clean electron–phonon coupled system insensitive to the random environment of the leads. To check this we isolate the double dot from the leads by symmetrically pinching-off its side barriers while maintaining a large internal tunnelling rate, $\Gamma_{\text{C}} \gg 2\pi f_i$ ($i = 1, 2$). Figure 5 shows the softening of the first and second modes measured in this regime along their effective gating directions ($V$ and $\varepsilon$ respectively) and plotted as a function of a normalized tunnelling rate to the leads, $\gamma = (\Gamma_{\text{L}} + \Gamma_{\text{R}})/2\pi f_i$. As the double dot is disconnected from the leads by lowering $\gamma$, we observe a clear quenching of the first-mode softening, in complete analogy with the observation for the single-dot case (Fig. 2). Remarkably, however, the detachment from the leads leaves the softening of the second mode essentially unaffected (evident by comparing the extreme cases $\gamma \ll 1$, and $\gamma > 1$, (top insets)). This intriguing observation can be understood by realizing that a double dot has an internal electronic degree of freedom, involving the transfer of charge between the dots, which provides the correlated electron flow that induces the softening (illustration, Fig. 5). This degree of freedom does not couple to the common-mode gating of the first mode but directly couples to the detuning gating of the second mode and thus softens only the latter, an effect that is nicely captured theoretically (Supplementary Information 8). The above measurements thus clearly demonstrate strong and selective coupling between phonons and internal electronic degrees of freedom in an isolated system.

The observation of tailored coupling between internal electronic and phononic degrees of freedom opens a wide range of new possibilities. One example pertains to the coupling of phonons to solid-state qubits, which use the singlet and triplet states of
two electrons in a double quantum dot as their basis.\textsuperscript{27,28} Owing to the Pauli blockade, an electron can shift between the dots only in the singlet state but not in the triplet state, and thus the second phonon frequency will dynamically couple only to the former. This selective coupling thus provides a tantalizing new route for transferring quantum entanglement from the electronic to the mechanical subsystems, or even between distant qubits in a multi-site lattice through a phonon ‘bus’.\textsuperscript{29,30} Generalizing the physics demonstrated here in double dots to multi-site lattices that are now well within reach\textsuperscript{22} would enable an even broader class of experiments that explore bulk electron–phonon phenomena, such as ferroelectricity, Peierls and Jahn–Teller instabilities, or superconductivity, in an engineered nanoscale setting. Analogous to the richness of quantum dot physics, made possible by the extensive control over their electronic properties, the ability to tailor the dynamics, spatial structure and selectivity of the coupling between electrons and phonons, demonstrated here, will enable the study of these phenomena in new regimes unattainable in bulk systems, opening new frontiers for fundamental experiments in condensed-matter physics.

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