Tunneling in suspended carbon nanotubes assisted by longitudinal phonons

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Current-voltage characteristics of suspended single-wall carbon nanotube quantum dots show a series of steps equally spaced in voltage. The energy scale of this harmonic, low-energy excitation spectrum is consistent with that of the longitudinal low-\(k\) phonon mode (stretching mode) in the nanotube. Agreement is found with a Franck-Condon-based model in which the phonon-assisted tunneling process is modeled as a coupling of electronic levels to underdamped quantum harmonic oscillators. Comparison with this model indicates a rather strong electron-phonon coupling factor of order unity.

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In nano-electromechanical systems (NEMS), mechanical motion affects electrical current and vice versa\[1, 2, 3, 4\]. Of special interest is the study of electron-phonon coupling in these devices since tunneling of a single electron may induce a displacement of the movable structure\[5, 6, 7, 8, 9, 10\]. The interaction strength is characterized by the dimensionless electron-phonon (e-ph) coupling constant \(g\), which is proportional to the ratio of the classical and the quantum displacement. In bulk systems the e-ph coupling is generally weak and the coupling constant is orders of magnitude smaller than one. However, since the coupling dramatically increases with decreasing device mass, NEM-devices may exhibit an intermediate to strong e-ph coupling\[11, 12, 13\]. In this regime, current-voltage characteristics are expected to exhibit additional steps whose height can be used as an estimate of \(g\). For example, \(g\) is around one in the \(C_{60}\) molecular devices of Ref.\[11\], while measurements on different \(C_{140}\) samples\[12\] indicate a value of \(g\) between 0.2 and 8.

Carbon nanotubes (NTs) are ideal systems for exploring electro-mechanical effects since they have a small diameter, a low mass, and can be defect free on a molecular level. In experiments on suspended nanotubes, different methods have already been used to probe the bending\[14, 15\] and radial breathing mode (RBM)\[16\]. The measurements show that the free-hanging tubes operate in the underdamped regime of low dissipation. For the fundamental bending mode the reported quality factor is about 100; for the RBM it is estimated as high as 10000.

In this Letter we present electronic transport spectroscopy measurements on suspended single-wall nanotubes, which show signatures of phonon-assisted tunneling, evidenced by the presence of a series of steps in the \(I - V\) characteristics. Such steps form a harmonic low-energy spectrum, whose energy scale and length dependence are consistent with that of the longitudinal stretching mode. Comparison with the Franck-Condon theory shows that the e-ph coupling constant is of order one.

Devices are fabricated by locating individual nanotubes (laser ablation and CVD) on a Si/SiO\(_2\) substrate using an atomic force microscope (AFM) with respect to predefined markers. Subsequently, the electrodes are made using conventional e-beam lithography techniques and thermal evaporation of Cr (5 nm) and Au (50 nm). The nanotubes are suspended by removing the underlying SiO\(_2\) in a wet etch step using buffered HF\[17\]. A schematic sample geometry and SEM micrograph are shown in Fig. 1. In the experiment the source and gate voltage are defined with respect to the drain, which is connected to ground.

In Fig. 2 we show stability diagrams for three nanotubes measured at 10 mK (a) and 300 mK (b,c) where the differential conductance, \(dI/dV\), is plotted versus bias and gate voltage. The three metallic nanotubes have a length between source and drain contacts, \(L\), ranging from 0.14 to 1.2 \(\mu\m\). Their diameter \(d\) is between 1 and 1.4 nm as determined from AFM imaging. In the diamond shaped regions (Coulomb diamonds) the current is zero due to Coulomb blockade, and the charge number in the dot is fixed. Regular and closing Coulomb diamonds indicate single dot behavior\[18, 19\] in all three samples for the gate range shown. Notice that the diamonds in Fig. 2a do close, as shown in the inset, which was taken at a higher temperature (300 mK) in a different cooldown. The low-bias current, however, is suppressed
which could be a signature of strong electron-phonon coupling [2, 3, 11].

Excitations of a quantum dot appear as lines running parallel to the Coulomb diamond edges in the stability diagrams [18]. At such a line, a new electronic level begins to develop. A new electronic level begins to develop. The energy of an excitation can be determined by reading off the intersection point between the excitation line and the Coulomb diamond edge, which we plot the mean electronic energy level separation, $\Delta$, in black. Blue squares correspond to the fundamental vibrational excitation energy extracted from the linear fits in the insets of Fig. 3. The energy of the radial breathing mode (green) does not depend on the nanotube length and equals $28 \text{meV} / \text{d} / \text{nm}$.

In Fig. 2a, we plot the energy of important low-energy vibrational modes of single-wall nanotubes [22, 23]. For comparison, we plot the mean electronic energy level separation, $\Delta$, in black. Blue squares correspond to the fundamental vibrational excitation energy extracted from the linear fits in the insets of Fig. 3. The energy of the radial breathing mode (green) does not depend on the nanotube length and equals $28 \text{meV} / \text{d} / \text{nm}$.

When the energy of an excitation is an integer multiple of the fundamental excitation energy, the excitation energy is equal to the electronic excitation energy, and the Franck-Condon principle predicts steps in the conductance. As a consequence, the transition rate is proportional to the product of the overlap integral between the vibrational wavefunctions and the density of states.

The energy of the radial breathing mode (green) does not depend on the nanotube length and equals $28 \text{meV} / \text{d} / \text{nm}$. The bending mode (red) has a $L^{-2}$ dependence [24] and an energy much smaller than the measured excitation energy. The stretching mode vibration energy (blue) is inversely proportional to the length [24], $E = (nh/L)\sqrt{Y/\rho_m}$, where $Y$ is Young’s modulus, $\rho_m$ is the density and $n$ is the vibrational quantum number. For nanotubes with $\rho_m = 1.3 \text{g/cm}^3$, $Y = 1 \text{TPa}$ the vibrational energy corresponding to the fundamental mode is $\sim 110 \text{meV} / L(\mu m)$ [24]. As Fig. 4 shows, the data are in good agreement with these predicted values.

The coupling of electronic levels with vibrational modes (quantum harmonic oscillators) can be described in terms of the Franck-Condon model [24]. According to the Franck-Condon principle, an electron in an electronic transition moves so fast that the nuclear positions are virtually the same immediately before and after the transition. As a consequence, the transition rate is proportional to the square of the overlap integral between the vibrational wavefunctions of the two states involved. An important parameter is the electron-phonon coupling factor, $g = \langle \frac{1}{2} (\frac{d}{dx})^2 \rangle$. This is the ratio of the classical displacement length, $x$, to the quantum mechanical oscillator length, $x_0 = \sqrt{\hbar / \omega m}$. Alternatively, $g = \frac{F^2}{2 \hbar \omega m}$, where $F$ is the force on, $m$ the mass of, and $\omega$ the frequency of the oscillator.

For low damping, the vibrational levels remain sharp and the Franck-Condon model predicts steps in the current-voltage characteristics, that are equally spaced in energy (bias voltage). In the presence of strong relaxation, the normalized step heights are given by [25]: $P_n = e^{-g n!} / n!$. In the strong coupling ($g \gg 1$) limit,
the height of the first steps is exponentially suppressed (phonon blockade) \[8\] \[10\]. Multiple steps only arise if \( g \) is of the order of one or larger and the observation of a spectrum of equally spaced excitation lines therefore indicates that the e-ph coupling in our suspended nanotubes must be rather strong.

In Fig. 3, the red curves represent the step heights \( P_n \) given by the Franck-Condon model with strong relaxation discussed above. The symbols are the experimental curves taken at the green lines in Fig. 2. Considering the simplicity of the model, reasonable agreement is obtained in all three cases. The comparison yields an estimate of \( g \) of 0.95, 1.1, and 0.5 in Fig. 3a, b, and c respectively, indicating that it is approximately length independent.

We have also performed a similar analysis at other gate voltages yielding the same \( g \)-values.

The theoretical curves in Fig. 3 do not exactly follow the measured ones. Better fits may be obtained if the influence of a gate voltage and asymmetric coupling is considered \[10\] or if coupling to excited electronic states \[24\] is considered or if the influence of damping or non-equilibrium phonons (weak relaxation) is taken into account. In the latter case the peak heights are expected to display a non-systematic dependence on \( g \) and peak number \( n \). Consideration of these effects is, however, beyond the scope of this paper.

The high value of the e-ph coupling \( (g \sim 1) \) is remarkably since in graphite the coupling between electrons and longitudinal phonons is weak. A source of intermediate to strong coupling could be the interaction between longitudinal and transverse vibrations \[28\]. Alternatively, we find that in suspended nanotubes the same e-ph coupling mechanism as in the bulk \[24\] can lead to a \( g \sim 1 \) if the electron density is inhomogeneous. The calculation proceeds as follows: The interaction energy of electrons with the polarization charge is characterized by the energy

\[
W = -\int dx dx' \rho(x) K(x-x') \frac{\partial P}{\partial x'}.
\]  

Here, \( \rho(x) \) is the density of excess charge produced by one electron, \( K(x-x') \) is an interaction kernel, which we approximate by \( \delta(x-x') \) for the case that interactions are effectively screened by the gate, and \( P(x) \approx e \rho_0 z(x) \) is the polarization vector. The quantity \( \rho_0 \sim k_F \) is the total electron density and \( z(x) \) is the displacement, which in the single-mode approximation becomes \( z(x) = A_n \sin(\pi n x/L) \). Calculating the force \( F = -\partial W/\partial A_n \), we obtain

\[
F = \frac{e \rho_0 \pi n}{L} \int_0^L dx \cos \frac{\pi n x}{L}.
\]  

If the excess charge density is uniform, \( \rho(x) = e/L \), \( F = 0 \) for all modes. Incorporation of interactions that are screened at distances longer than the distance to the gate, yields a force that scales as \( L^{-2} \). In this case, the coupling
parameter $g$ also scales as $L^{-2}$ and typical values are in the order of $10^{-3}$, in apparent contradiction with the experimental data.

Assuming that the charge is localized in the center of the tube, $\rho(x) = e\delta(x - L/2)$, the force is zero for odd harmonics, but for even harmonics, $n = 2l$, it reads $F_l = (-1)^l e^2 \rho_0 2\pi l / L$. This results in a coupling parameter that is length independent and scales as $l^{-1}$: higher modes are coupled weaker to electrons. Numerical estimates show that $g \sim 1$. Localization of an electron in a point away from the middle produces coupling to both odd and even modes. Note that the electron does not have to be strongly localized to produce a $g \sim 1$. Such a non-uniform density can be created by impurities located in the substrate, or induced by a redistribution of electrons in a suspended tube bent by a underlying gate electrode.

An interesting feature of the data is the appearance of negative differential conductance (NDC) in the current-voltage characteristics. NDC is very pronounced in suspended nanotube quantum dots whose heights are in reasonable agreement with the Franck-Condon predictions if the e-ph coupling constant is of order unity. Suspended nanotube quantum dots showed signatures of phonon-assisted tunneling, mediated by longitudinal vibrational (stretching) modes. The current-voltage characteristics show multiple steps and PDC lines may have the same gate voltage dependence preceded by a region of suppressed current. This scenario may especially be relevant for the data in Fig. 3a, but also present in Fig. 3b,c. Although several explanations for NDC have been put forward, its origin remains unclear. Koch and von Oppen showed that for low relaxation and strong e-ph coupling, NDC features appear, although they do not follow regions with strong positive differential conductance (PDC) as in our data. McCarthy et al.

In summary, transport measurements on suspended SWNTs show signatures of phonon-assisted tunneling, mediated by longitudinal vibrational (stretching) modes. The current-voltage characteristics show multiple steps whose heights are in reasonable agreement with the Franck-Condon predictions if the e-ph coupling constant is of order unity. Suspended nanotube quantum dots form an interesting model system for future studies on the interaction between single electrons and quantized phonons in the intermediate to strong electron-phonon coupling limit.

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