Novel spectral features of nanoelectromechanical systems

M. Tahir1, A. MacKinnon2 & U. Schwingenschlögl1

1PSE Division, KAUST, Thuwal 23955-6900, Kingdom of Saudi Arabia, 2Department of Physics, The Blackett Laboratory, Imperial College London, South Kensington Campus, London SW7 2AZ, United Kingdom.

Electron transport through a quantum dot or single molecule coupled to a quantum oscillator is studied by the Keldysh nonequilibrium Green’s function formalism to obtain insight into the quantum dynamics of the electronic and oscillator degrees of freedom. We tune the electronic level of the quantum dot by a gate voltage, where the leads are kept at zero temperature. Due to the nonequilibrium distribution of the electrons in the quantum dot, the spectral function becomes a function of the gate voltage. Novel spectral features are identified for the ground and excited states of nanomechanical oscillators that can be used to enhance the measurement sensitivity.

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NEMS have become a promising prototype for measuring the quantum behavior of nanoscale objects, theoretically as well as experimentally. The main motivation to study NEMS is the large number of potential applications in nanoelectronic devices, such as the measurement of very small masses, weak forces, and ultra small displacements of atomic positions. NEMS are also important from a fundamental point of view for understanding the transition between the classical and quantum regimes. This includes the observation, measurement, and control of quantum states of mesoscopic mechanical continuous variable systems, such as the quantum harmonic oscillator. Metallic leads coupled to the quantum dot have been successfully used to characterize the state of the mechanical resonator classically and quantum mechanically. When the device is scaled down to the nanometer regime, the resonance frequencies increase and at the same time the effective masses decrease. Related applications are discussed in a very recent review in Ref. 20. Various electromechanical properties of the devices, such as current, conductance, and shot noise, are proportional to the spectral function. Hence, the spectral features of NEMS have not been discussed so far.

In view of the above, we study in this work the spectral function of a single quantum dot connected to two metallic leads and a quantum harmonic oscillator. An arbitrary voltage is applied to tune the single electronic level of the dot. The quantum oscillator is in the ground state and the leads are assumed to be in equilibrium at zero temperature. We address very small to very large chemical potential differences between the Fermi levels of the left and right leads, where the response to both Fermi levels is included in the calculations. The position of the resonant level with respect to the chemical potential in the leads is thus affected by the displacement of the nanomechanical oscillator, which in turn affects the transport properties of the tunnel junction. Electronic transport properties of similar systems have been discussed for a single electron transistor and a superconducting single electron transistor, both coupled to an oscillator. Both these studies have employed the quantum master equation approach within the weak coupling limit. It has been observed that, even though the leads are at zero temperature, the oscillator behaves as if it would be coupled to a bath/reservoir of finite temperature related to the chemical potential difference.
Here we employ the nonperturbative Green’s function approach\textsuperscript{19,23}, as the recent experiments go beyond perturbation theory and thus a nonperturbative approach to the quantum master equation (equation of motion) is required. The Green’s function approach allows us to consider strong electron-phonon coupling and has been applied successfully for very small up to very large applied bias. In contrast, the equation of motion approach\textsuperscript{14,25,26} treats both couplings as perturbations. In addition, this approach is valid only for high applied bias. We assume that quantum transport through the tunnel junction can be described by sequential tunneling. We note that the electron-oscillator interactions have been studied by scanning tunneling microscopy\textsuperscript{36,37,27–29} and spectroscopy\textsuperscript{29,30}. These methods turn out to be more valuable tools than the inelastic tunneling spectroscopy\textsuperscript{31–33}. In Refs. 34–39 it has been shown that the position of the single quantum dot energy level and the tunneling width, \( \Gamma \), strongly affect the Kondo spectrum, where a third terminal has been used as a tip detector to measure the Kondo peaks. Spectral features of the Kondo effect have also been observed in the presence of strong electron-phonon interaction\textsuperscript{40–42}. In the present work, therefore, we will consider whether the intriguing features of NEMS can be explored by the spectral function.

**Model formulation and the spectral function**

We consider a single quantum dot connected to two identical metallic leads and apply a gate voltage. The single mode of the quantum oscillator is strongly coupled to the electrons on the dot. We neglect the spin degree of freedom, the electron-electron interaction, the effects of finite electron temperature of the leads and the damping of the oscillator. The Hamiltonian \( H = H_{\text{dot-ph}} + H_{\text{leads}} + \sum_{\eta=L,R} H_{\eta,\text{dot}} \) to be studied in the following is thus given by\textsuperscript{19,21–24}

\[
H_{\text{dot-ph}} = \left[ E_0 + \lambda (b^\dagger b + b^\dagger b) \right] c_0^+ c_0 + \Omega b^\dagger b, \tag{1}
\]

where \( E_0 \) is the energy of the single electron level on the dot and \( c_0^+ \) and \( c_0 \) are the corresponding creation and annihilation operators. Moreover, \( \lambda \) is the coupling strength between the electrons on the dot and an oscillator of frequency \( \Omega \) (setting \( \hbar = 1 \) for simplicity) and \( b^\dagger, b \) are the raising and lowering operators of the phonons. The remaining elements of the Hamiltonian are

\[
H_{\text{leads}} = \sum_{\eta=L,R} \sum_{j=1}^{N} E_{\eta,j} c_{\eta,j}^+ c_{\eta,j}, \tag{2}
\]

\[
H_{\eta,\text{dot}} = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} V_\eta \left( \frac{b^\dagger c_{\eta,j} c_{\eta,j}^+ + c_{\eta,j} c_{\eta,j}^+ b^\dagger}{\sqrt{N}} \right), \tag{3}
\]

where \( N \) is the total number of states in the lead and \( j \) counts the channels. We use the usual harmonic oscillator eigenfunctions \( \Psi_n(x/\ell) \) when the electron is in the lead and \( \Psi_n(x-x_\eta)/(\ell) \) when it is on the dot, where \( \ell \) is the zero point amplitude of the oscillator and \( x_\eta = \ell \lambda/\Omega \) is the displacement of the oscillator equilibrium position due to the coupling to the electrons. The Hamiltonian thus takes the form

\[
H' = \begin{pmatrix}
H'_L & V'_L & 0 \\
V'_L & H'_{\text{dot-ph}} & V'_R \\
0 & V'_R & H'_R
\end{pmatrix}, \tag{4}
\]

where \( H'_{\text{dot-ph}} = [E_0 + (n+1/2)\Omega - \Delta] \delta_{n,n'} \delta_{\ell,\ell'} \), \( H'_\eta = [E_{\eta,j} + (n+1/2)\Omega] \delta_{\eta,n} \delta_{\ell,\ell'} \), and \( \Delta = \ell^2/2\Omega \) is an energy shift.

In order to transform between the representations for the occupied and unoccupied dot we require the matrix elements

\[
M_{m,n} = \ell^{-1} \int \Psi_m^*(x-x_\eta)/(\ell) \Psi_n(x/\ell) \, dx \tag{5}
\]

which may be simplified\textsuperscript{43} as

\[
M_{m,n} = \sqrt{\frac{2^{m-n} \min(m,n)!}{\max(m,n)!}} \exp\left(-\frac{1}{4} x^2\right) \left(\frac{i}{2}\right)^{n-m} L_{\min(m,n)}^{\max(m,n)} \left(\frac{1}{2} x^2\right), \quad \tag{6}
\]

where \( x = x_\eta/\ell = \lambda/\Omega \). In addition, \( L_n^m(x) \) are the associated Legendre polynomials.

The self-energy represents the contribution to the dot energy due to interaction between the dot and the leads it is coupled to. In the current model, with a single electronic state on the dot, there is no self-energy due to interaction. We use the wide-band approximation, where the self-energy contribution of each lead becomes energy independent and can be calculated as

\[
\Sigma^\tau_{n,n,\eta} (\omega) = H^\tau_{\eta,\text{dot}} \Gamma^\tau_{n,n,\eta} (\omega - \omega - (n+\frac{1}{2})\Omega), \tag{7}
\]

where

\[
g^\tau_{n,\eta} (\omega - (n+\frac{1}{2})\Omega) = \frac{1}{N} \int_{-\infty}^{+\infty} \frac{\rho_\eta dE_n}{\omega - (n+\frac{1}{2})\Omega - E_n}, \tag{8}
\]

\( \rho_\eta \) is the density of states in lead \( \eta \) (L or R), and \( g^\tau_{n,\eta} (\omega) \) is the uncoupled retarded Green’s function in the leads (recall that \( \hbar = 1 \)). The retarded self-energy can hence be written as

\[
\Sigma^\tau_{n,n,\eta} (\omega) = -i \frac{1}{2} \Gamma_\eta. \tag{9}
\]

Here \( \Gamma_\eta = 4\pi |V_\eta|^2 \rho_\eta \) and \( \Gamma_\eta \) is the damping factor (\( \Gamma_L = \Gamma_R = \Gamma \)). The retarded self-energy is now independent of the oscillator index. Similarly

\[
\Sigma^a_{n,n,\eta} = \left[ \Sigma^\tau_{n,n,\eta} \right]^* = +i \frac{1}{2} \Gamma_\eta. \tag{10}
\]

We solve Dyson’s equation using \( H_{\eta,\text{dot}} \) as a perturbation. In the presence of the oscillator, the retarded and advanced Green’s functions on the dot, with the phonon states in the representation of the unoccupied dot, may be written as

\[
G^a_{n,m} (\omega) = \sum_{m'} M_{m,m'} G^a_{m,m'} (\omega) M^\dagger_{n,m'}, \tag{11}
\]

where \( G^a_{n,m}(\omega) \) is the retarded/advanced Green’s function on the occupied dot obtained as

\[
g^a_{n,\eta}(\omega) = \left[ \omega - E_0 - \left( \frac{m+\frac{1}{2}}{2} \right) \Omega + \Delta \pm i\Gamma \right]^{-1}. \tag{12}
\]

The lesser Green’s function in the presence of the nanomechanical oscillator including the dot and the leads is written as

\[
G^-_{n,\eta} (\omega) = \sum_{m,n_\eta} G^-_{m,n_\eta} (\omega) \Sigma^-_{m,n_\eta} (\omega) G^a_{n,m_\eta}(\omega). \tag{13}
\]

Here \( \Sigma^- (\omega) \) is the lesser self-energy, given by

\[
\Sigma^- (\omega) = \Sigma^-_L (\omega) + \Sigma^-_R (\omega), \tag{14}
\]

where the off-diagonal elements of the matrix \( \Sigma^- (\omega) \) are zero and the diagonal elements may be written as

\[
\Sigma^-_{n,n_\eta} (\omega) = i\hbar \int dE f_{\eta} (E_\eta) B_n (\omega - E_\eta - (n+\frac{1}{2})\Omega), \tag{15}
\]

where \( \epsilon = \omega - (n+\frac{1}{2})\Omega \) and \( f_\eta (E_\eta) \) denotes the Fermi distribution functions of the left and right leads, which have different chemical potentials under the bias, and \( B_n \) is the Boltzmann factor of the oscillator state. The index \( n \) determines the statistical occupation.
probability of the phonon state $|n\rangle$ at finite temperature. Therefore, the accessibility of particular conduction channels is determined by a weight factor of the Boltzmann distribution function. For the present case of zero temperature the lesser self-energy may be recast in terms of the Heaviside step function $\theta(x)$ as

$$\sum_{n,n'=0}^{\infty} \langle n|\theta\left(E_{F_{\eta}} + \frac{1}{2} \Omega - \omega\right) \delta_{n,n'},$$

where $E_{F_{\eta}}$ is the Fermi energy on lead $\eta$ and the Kronecker delta, $\delta_{n,n'}$, signifies that the nanomechanical oscillator is initially in its ground state, $n = 0$. All physical properties may be expressed in terms of the spectral function $A(\omega)$, which is at the heart of this paper. The spectral function is related to the imaginary part of the retarded or advanced Green’s function$^{21,22,25}

$$A(\omega) = - \text{Im}[G^r(\omega)] = + \text{Im}[G^a(\omega)],$$

which we obtain as

$$A(\omega) = \sum_{n} \frac{2\pi M_{n,n} M_{n,n}^*}{|\omega - E_{0} - \left(m + \frac{1}{2}\right)\Omega + \Delta|^2 + \Gamma^2},$$

where $n = n' = 0$ for the ground state and $1$ for the first excited state of the nanomechanical oscillator. The spectral function is then used to calculate the differential conductance via the relation

$$I = \frac{e^2}{4\pi} \int d\omega \lambda(\omega) [f_L(\omega) - f_R(\omega)],$$

where we have fixed the dot energy to $E_0 = 0.5$ and set the Fermi energy of the right lead to zero. The spectral function is proportional to the density of electron plus phonon states by the relation $n(\omega) = A(\omega)/\pi$, which gives information about the excitations (electron or hole) of the system. The spectral function obtained in Eqs. (17) to (19) is the main result of the present work and the starting point for the subsequent discussion. The form of the spectral function obtained here makes the electronic and oscillator properties apparent as it is the essential ingredient for theoretical considerations on problems such as the high frequency behavior of nanomechanical oscillators, the current-voltage characteristics, quantum conductance, and shot noise phenomena$^{21-25,34-42}$.

**Discussion**

Analytical examination of Eq. (17) demonstrates a resonant peak for the purely electronic degree of freedom plus any number, $m$, of phonons. This discrete structure confirms the quantum mechanical aspect of the calculation$^{18,26}$. We expect the side peaks to be suppressed in the absence of electron-phonon coupling through $M_{m,n} = 0$ for $m \neq n$. In this regime, the resonant peak is shifted in energy by a constant shift, $\Delta$, as shown in Eq. (17). In all the subsequent figures the following parameters are used with the dimension of energy: $\Omega = 0.3$, $E_0 = 0.5$, $\lambda = 1.1\Omega$, and $\Gamma = 0.2\Omega$.

We discuss the spectral function in Fig. 1 for the ground state ($n = 0$, dotted line) and the first excited state ($n = 1$, solid line) of the nanomechanical oscillator. We find $n$ side peaks and a constant shift $\Delta$ consistent with Eqs. (4) and (17). This is the regime of strong coupling, $\lambda = 1.1\Omega$, whereas in the limit of weak coupling, $\lambda = 0.5\Omega$, we find a single side peak for the excited state. The amplitude of the side peaks becomes larger and the peaks sharper when the oscillator starts in the excited state instead of the ground state. We find a nontrivial feature for the first excited state of the nanomechanical oscillator, see the solid lines in Fig. 1: The first side peak is suppressed with a very small amplitude as compared to all other side peaks for strong electron-phonon coupling, which is not the case when the nanomechanical oscillator starts in its ground state (dotted lines in Fig. 1).

In order to explore this nontrivial behavior, we show in Figs. 2 and 3 three-dimensional plots of the spectral function, as a function of $\omega$ and $\lambda$, for the ground state ($n = 0$) and excited ($n = 1$) states, respectively. We observe that valleys appear when the values of the excited state and integer values coming from the probability density function in Eq. (17) are the same. Closer examination of this equation shows that for $m = 1$ the side peaks are suppressed for different coupling strengths. Such a behavior is not possible in classical dynamics$^{18,26}$ or any other perturbative approach used for molecular systems$^{21-25}$. Finally, we show the probability density $P(x) = M_{m,n} M_{n,m}^*$ calculated by Eq. (6) in Fig. 4 as a function of the...
dimensionless coupling strength $x$ to confirm the suppression of the side peaks. Results for the ground state and the first excited state of the nanomechanical oscillator are given by dotted and solid lines, respectively. From the solid line it is clear that when the oscillator starts in an excited state the curve approaches zero for $n$ values of the coupling. Using Eq. (6), we obtain for the probability

$$P(x) \propto \left[ \frac{1}{2} (1 - x^2) \right]^2,$$

which can be simplified using for $x > 0$ the identity

$$L_m^m(x) = \frac{1}{\sqrt{\pi}} e^{x^2/2} x^{-(2m+1)/4} n^{(2m-1)/2} \cos \left( 2 \sqrt{n} x - \frac{(2m+1)\pi}{4} \right) + O \left( n^{(2m-3)/4} \right)$$

to yield

$$P(x) \propto \left[ \frac{1}{\sqrt{\pi}} e^{x^2/4} (x^2/2)^{-1/4} \cos \left( 2x - \frac{\pi}{4} \right) \right]^2.$$

No such behavior has been reported for classical and perturbation treatments. The reason is that in the classical mechanics and perturbation approaches the oscillator does not couple strongly enough to the electrons on the dot. The nontrivial behavior only appears because a nonperturbative approach has been used in the present work. As compared to the existing literature, our nonperturbative approach is valid for small to large applied bias and can be employed for studying various problems in molecular systems, NEMS, and shuttling transport phenomena. The theoretical framework developed in the present work for describing the ground and excited states of NEMS provides both reliable predictions of experiments and improved understanding of the circumstances under which nanomechanical systems are governed by quantum behavior.

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**Author contributions**

M.T., A.M. and U.S. wrote the manuscript.

**Additional information**

Competing financial interests: The authors declare no competing financial interests.

**How to cite this article:** Tahir, M., MacKinnon, A. & Schwingschlägl, U. Novel spectral features of nanoelectromechanical systems. *Sci. Rep.* **4**, 4035; DOI:10.1038/srep04035 (2014).

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