Temperature dependence of electron transport through a quantum shuttle

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We analyze electron transport through a quantum shuttle with an applied voltage below the instability threshold. In this, we determine the current-voltage characteristics of the system and show that at low temperatures they exhibit pronounced steps. The temperature dependence of the current is calculated in the range from 2 K to 300 K and it is seen to demonstrate a wide variety of behaviors—from $1/T$ decreasing to an exponential growth—depending on how deep the shuttle is in the quantum regime. Our results are compared to experimental data on electron transport through long molecules.

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Recent achievements in nanotechnology facilitate the development of a new generation of nanodevices incorporating mechanical degrees of freedom, nanoelectromechanical systems (NEMS).[1–3] In this, one of the most promising device structures is a single-electron shuttle[4] carrying an electron flow between two contacts. Even classical aspects of this model were shown to be important for a description of electron transport through a nanomechanical bell[5] and a C_{60} single-molecule transistor.[6] Theoretical analysis of such a system in the classical framework was done in Refs. 7–9. However, many properties of NEMS can only be understood on a fully quantum-mechanical basis. The quantum regime is reached for a shuttle (with mass m and characteristic frequency ω_0) when the amplitude of zero-point mechanical fluctuations, $\Delta x_{zp}=\sqrt{\hbar/2m\omega_0}$, is of the order of a tunneling length λ [involved in dependence of the tunnel matrix elements on the distance x between the shuttle and the leads, $T_{k\alpha}(x)=T_{k\alpha}\exp(x/\lambda_\alpha)$, $\alpha=L,R;\lambda_L=-\lambda,\lambda_R=\lambda$]. The effects of quantization of the mechanical motion were examined in Refs. 10–12, and the occurrence of shuttle instability was predicted in Refs. 13,14 for the zero-temperature case.

In the present paper we examine electron transport through the shuttle system at low applied bias voltage (below the instability threshold), with emphasis on the temperature effects. To accomplish this, we perform a theoretical analysis encompassing both the mechanical motion and electron transport using a model of the quantum shuttle with a single-electron level (strong Coulomb blockade regime). This approximation is valid, in particular, for the C_{60} shuttle system, when the charging energy exceeds 270 meV,[6] which is much larger than all other energetic parameters involved. We show that at low temperature the current-voltage characteristics exhibit pronounced steps. The first step takes place when the applied voltage just compensates the initial separation between the single-electron energy level in the shuttle and the equilibrium chemical potential in the leads. The second step occurs when the applied voltage facilitates electron tunneling from the left lead to the shuttle with the absorption of a virtual quantum of mechanical motion (‘‘phonon’’) by the electron, with subsequent emission of this phonon during electron tunneling from the shuttle to the right lead. Moreover, we demonstrate that there is instability of electron transport through the shuttle when the applied voltage is sufficient to produce a real phonon absorbed by the shuttle system. With further increase in voltage, the oscillator becomes anharmonic and new nonlinear effects prevent the increase in infinite current. However, the examination of this situation is beyond the scope of the present paper and will be performed elsewhere.

We show that the behavior of the temperature dependence of the current through the system is strongly dependent on the quantum parameter

$$\nu_0=\frac{\hbar}{2m\omega_0\lambda^2},$$

which describes the level of zero-point mechanical fluctuations relative to the tunneling length. In particular, we demonstrate that systems with relatively small zero-point mechanical fluctuations ($\nu_0\leqslant 0.1$) exhibit weak temperature dependence of the current-voltage characteristics $I(V,T)$, whereas for the quantum shuttle with $\nu_0\geqslant 0.4$ we obtain the current as an exponential function of temperature. It should be emphasized that the model of the quantum shuttle can be important in the theoretical explanation of some aspects of electron transport through single molecules and self-assembled monolayers.[15] An organic molecule is coupled to leads with elastic links[16] and may oscillate as a whole. These oscillations are reflected in the temperature dependence of the current-voltage characteristics of the system.[17] It should be noted that experimental current-voltage characterizations of organic monolayers demonstrate a wide variety of temperature behaviors—from a weak temperature dependence for molecules C8, C12, and C16, sandwiched between Au contacts,[18] to an exponential temperature dependence of conductance and current that has been observed by Stewart *et al.*[19] in experiments with a Langmuir-Blodget monolayer of eicosanoic acid (C20) connected to Pt electrodes. The latter results have not yet been explained with well-known mechanisms of molecular transport (direct tunneling or electron hopping) because no temperature dependence is expected in the case of direct tunneling, whereas for the hopping mechanism temperature behavior of the current and conductance should be described by the activation dependence. We suggest that mechanical molecular motion may be the reason for such an exponential dependence. The difference in measured temperature dependencies may be attributed to the difference in connections of the molecules to electrodes and, correspondingly, to the different parameters ν_0 for these systems.

To examine electron transport through the quantum shuttle, we start from a Hamiltonian having the form ($\hbar=1$, $k_B=1$)

$$H=\frac{p^2}{2m}+\frac{m\omega_0^2x^2}{2}+E_0a^+a+\sum_{k\alpha}E_{k\alpha}c_{k\alpha}^+c_{k\alpha}-\sum_k(T_{k\alpha}c_{k\alpha}^+a+T_{k\alpha}^*a^+c_{k\alpha})e^{x/\lambda_\alpha}. \quad (1)$$

Here $c_{k\alpha}^+/c_{k\alpha}$ are creation/annihilation operators of electrons in the α lead, a^+/a are creation/annihilation operators of electrons in the shuttle, and V is the voltage applied to the leads.[20] The equations of motion, derived from the Hamiltonian, (1), are given by

$$i\dot{a}=E_0a-\sum_{k\alpha}T_{k\alpha}^*c_{k\alpha}e^{x/\lambda_\alpha},$$

$$i\dot{c}_{k\alpha}=E_{k\alpha}c_{k\alpha}-T_{k\alpha}ae^{x/\lambda_\alpha},$$

$$\ddot{x}+\omega_0^2x=\sum_{k\alpha}\frac{1}{m\lambda_\alpha}(T_{k\alpha}c_{k\alpha}^+a+T_{k\alpha}^*a^+c_{k\alpha})e^{x/\lambda_\alpha}. \quad (2)$$

It should be noted that we take into account nonlinearity associated with the nonlinear exponential dependence of the tunneling elements on the distance between the shuttle and the leads.

The electric current through the shuttle is defined as $I=I_L=-I_R$, where

$$I_\alpha=e\frac{d}{dt}\sum_k\langle c_{k\alpha}^+c_{k\alpha}\rangle=-ie\sum_k T_{k\alpha}^*\langle a^+e^{x/\lambda_\alpha}c_{k\alpha}\rangle+\text{H.c.} \quad (3)$$

With the expression for the electron operator in the lead derived from Eq. (2) as

$$c_{k\alpha}(t)=c_{k\alpha}^{(0)}(t)+iT_{k\alpha}\int dt_1e^{-iE_{k\alpha}(t-t_1)}a(t_1)e^{x(t_1)/\lambda_\alpha}\times\theta(t-t_1), \quad (4)$$

where $\theta(t-t_1)$ is the unit step function, we obtain the electric current as

$$I_\alpha=-e\sum_k|T_{k\alpha}|^2\int dt_1f_\alpha(E_{k\alpha})e^{-iE_{k\alpha}(t-t_1)}\theta(t-t_1)\langle[a^+(t)e^{x(t)/\lambda_\alpha},a(t_1)e^{x(t_1)/\lambda_\alpha}]_+\rangle$$
$$+e\sum_k|T_{k\alpha}|^2\int dt_1e^{-iE_{k\alpha}(t-t_1)}\theta(t-t_1)\langle a^+(t)a(t_1)\rangle\langle e^{x(t)/\lambda_\alpha},e^{x(t_1)/\lambda_\alpha}\rangle+\text{H.c.} \quad (5)$$

To derive this expression, we have employed the formula

$$\langle a^+(t)e^{x(t)/\lambda_\alpha}c_{k\alpha}^{(0)}(t)\rangle=\int dt_1\langle c_{k\alpha}^{(0)+}(t_1)c_{k\alpha}^{(0)}(t)\rangle\left\langle\frac{\delta(a^+(t)e^{x(t)/\lambda_\alpha})}{\delta c_{k\alpha}^{(0)+}(t_1)}\right\rangle$$
$$=-iT_{k\alpha}\int dt_1\langle c_{k\alpha}^{(0)+}(t_1)c_{k\alpha}^{(0)}(t)\rangle\theta(t-t_1)\langle[a^+(t)e^{x(t)/\lambda_\alpha},a(t_1)e^{x(t_1)/\lambda_\alpha}]_+\rangle$$
$$=-iT_{k\alpha}\int dt_1f_\alpha(E_{k\alpha})e^{-iE_{k\alpha}(t-t_1)}\theta(t-t_1)\langle[a^+(t)e^{x(t)/\lambda_\alpha},a(t_1)e^{x(t_1)/\lambda_\alpha}]_+\rangle, \quad (6)$$

which can be understood from the fact that the product $a^+(t)e^{x(t)/\lambda_\alpha}$ is an operator functional of unperturbed operators of the leads $c_{k\alpha}^{(0)}(t),c_{k\alpha}^{(0)+}(t)$. These operators describe a free electron gas in the leads with the correlation function $g_{k\alpha}^<(t,t_1)=i\langle c_{k\alpha}^{(0)+}(t_1)c_{k\alpha}^{(0)}(t)\rangle=if_\alpha(E_{k\alpha})e^{-iE_{k\alpha}(t-t_1)}$, where $f_\alpha(E)=f(E-\mu_\alpha)=\{\exp[(E-\mu_\alpha)/T]+1\}^{-1}$ is the Fermi distribution. In order to eliminate the unperturbed operators, we have to pair the operator $c_{k\alpha}^{(0)}(t)$ of the lefthand side of Eq. (6) with the operator $c_{k\alpha}^{(0)}(t_1)$ involved in the functional and multiply this product by the functional derivative. This derivative can be calculated as the anticommutator of the operator $a^+(t)e^{x(t)/\lambda_\alpha}$ with the operator $a(t_1)e^{x(t_1)/\lambda_\alpha}$ conjugated to $c_{k\alpha}^{(0)+}(t_1)$ in the Hamiltonian (1) and in the corresponding S matrix. The shuttle electron correlators are approximately given by $\langle a^+(t)a(t_1)\rangle=Ne^{iE_0(t-t_1)}$, $\langle a(t_1)a^+(t)\rangle=(1-N)e^{iE_0(t-t_1)}$ with $N=\langle a^+a\rangle$ being the steady-state electron population of the shuttle, which can be determined from the condition $I_R+I_L=0$. Using previously derived formulas (see Ref. 17, Appendix) in the wide-band limit we obtain the expression for the current as

$$I_\alpha=e^{\nu_c}e\Gamma_\alpha\sum_{m=-\infty}^{+\infty}\sum_{l=-\infty}^{+\infty}J_m(\nu_0)I_l(\nu_c)\times\{N[1-f_\alpha(E_0-m\omega_0-l\omega_0)]-(1-N)f_\alpha(E_0+m\omega_0+l\omega_0)\}. \quad (7)$$

Here, $J_m(\nu_0)$ and $I_l(\nu_c)$ are the ordinary and modified Bessel functions of order m and l, respectively, $\Gamma_\alpha = 2\pi\Sigma_k|T_{k\alpha}|^2\delta(\omega-E_{k\alpha})$, and $\nu_c=\langle\tilde{x}^2\rangle/\lambda^2$ is the dispersion of mechanical oscillations of the nonequilibrium case relative to λ^2. In the following, we assume symmetric coupling between the shuttle and the leads, $\Gamma_L=\Gamma_R=\Gamma$. The steady-state value of the electron population in the shuttle is determined by the relation

$$N=C/D, \tag{8}$$

with

$$C=\sum_{ml} J_m(\nu_0)I_l(\nu_c)[f_L(E_0+m\omega_0+l\omega_0) + f_R(E_0+m\omega_0+l\omega_0)]$$

and

$$D=\sum_{ml} J_m(\nu_0)I_l(\nu_c)[2-f_L(E_0-m\omega_0-l\omega_0) + f_L(E_0+m\omega_0+l\omega_0)-f_R(E_0-m\omega_0-l\omega_0) + f_R(E_0+m\omega_0+l\omega_0)]. \tag{9}$$

For the case of relatively small zero-point mechanical fluctuations, $\nu_0 \ll 1, \Sigma_l I_l(\nu_c)=e^{\nu_c}$, we obtain simple formulas for the shuttle population N and for the current I as

$$N=e^{-\nu_c}\sum_l I_l(\nu_c)\frac{f_L(E_0+l\omega_0)+f_R(E_0+l\omega_0)}{2},$$

$$I=I_L=e\Gamma e^{\nu_c}\sum_l I_l(\nu_c)\frac{f_R(E_0+l\omega_0)-f_L(E_0+l\omega_0)}{2}. \tag{10}$$

It should be noted that the effective temperature of the *biased* nanomechanical system, which is determined by the dispersion of mechanical fluctuations $\langle\tilde{x}^2\rangle$, can be different from the equilibrium temperature T,[17] so one cannot neglect nonlinearities caused by ν_c even when $\nu_0 \ll 1$.

To calculate the relative dispersion of mechanical fluctuations, $\nu_c=\langle\tilde{x}^2\rangle/\lambda^2$, Eq. (2) for the shuttle position can be rewritten in the form of a quantum Langevin equation

$$\ddot{x}+\gamma\dot{x}+\omega_0^2 x=\xi, \tag{11}$$

with a damping rate

$$\gamma=\nu_0\Gamma e^{\nu_c}B(\omega_0) \tag{12}$$

and a fluctuation source

$$\xi=\sum_{k\alpha}(T^*_{k\alpha}/m\lambda_\alpha)\{a^+e^{x/\lambda_\alpha},c^{(0)}_{k\alpha}\}+\text{H.c.}, \tag{13}$$

which is characterized by the spectrum $K(\omega) =\nu_0 e^{\nu_c}\Gamma(\hbar\omega_0/m)A(\omega_0)$. Here, we have introduced $A(\omega)$ and $B(\omega)$ as the following dimensionless combinations of Fermi distribution functions:

$$A(\omega)=\sum_\alpha\sum_{ml} J_m(\nu_0)I_l(\nu_c)\times\{N[2-f_\alpha(E_0-m\omega_0-l\omega_0 -\omega)-f_\alpha(E_0-m\omega_0-l\omega_0+\omega)]+(1-N)[f_\alpha(E_0 +m\omega_0+l\omega_0+\omega)+f_\alpha(E_0+m\omega_0+l\omega_0-\omega)]\},$$

$$B(\omega)=\sum_\alpha\sum_{ml} J_m(\nu_0)I_l(\nu_c)\{N[f_\alpha(E_0-m\omega_0-l\omega_0-\omega) -f_\alpha(E_0-m\omega_0-l\omega_0+\omega)] +(1-N)[f_\alpha(E_0+m\omega_0+l\omega_0-\omega) -f_\alpha(E_0+m\omega_0+l\omega_0+\omega)]\}. \tag{14}$$

The relative level of mechanical fluctuations, ν_c, can be found self-consistently from the equation

$$\nu_c=\nu_0\frac{A(\omega_0)}{B(\omega_0)} \tag{15}$$

supplemented by the Eq. (8) for the steady-state shuttle electron population N.

We have solved a self-consistent set of equations, Eqs. (8) and (15), and substituted the obtained values of N and ν_c into Eq. (7) for electric current. The resulting current-voltage characteristics are shown in Fig. 1 for various temperatures, with energy separation $E_0-\mu=0.2\omega_0$, $\omega_0/2\pi=1$ THz, and with quantum parameters $\nu_0=0.7$ [Fig. 1(a)] and $\nu_0=0.07$ [Fig. 1(b)]. It is evident from these figures that there are pronounced steps in the low-temperature current-voltage characteristics at the bias voltages (i) $eV/2=E_0-\mu$ and (ii) $eV/2=\omega_0-E_0+\mu$. The first step occurs when the chemical potential of the left lead passes through the energy level of the shuttle, while the second one corresponds to the passing of the chemical potential of the right lead through the virtual level having energy $E_0-\omega_0$. In the latter case, tunneling of an electron of the left lead having energy $E_0-\omega_0$ to the shuttle level with energy E_0 is accompanied by the absorption of virtual quantum of mechanical motion (phonon), with further tunneling of this electron to the state of the right lead having energy $E_0-\omega_0$ accompanied by the emission of this phonon. This second step is much more pronounced for the case of larger ν_0. When the chemical potential of the left lead passes through the virtual level with energy $E_0+\omega_0$ (at the voltage $eV/2=\omega_0+E_0-\mu$), an electron tunneling from the left lead to the shuttle can be accompanied by the absorption of a real phonon by the shuttle, and the system becomes unstable. To illustrate this, we present, in the inset of Fig. 1(a), the current-voltage characteristics (logarithmic scale) for various values of $E_0-\mu$. It should be noted that the condition for this instability is identical to that found in Ref. 14. Finally, one can see in Fig. 11(a) that the current-voltage characteristics become smoother and the steps disappear with increasing temperature.

The temperature dependence of the current through the quantum shuttle system is of special interest. It is exhibited in Fig. 2 (logarithmic scale) for $E_0-\mu=0.2\omega_0$, $eV =0.4\omega_0$, $\omega_0/2\pi=1$ THz, with various values of quantum parameter ν_0. It is evident from this figure that an initial $1/T$

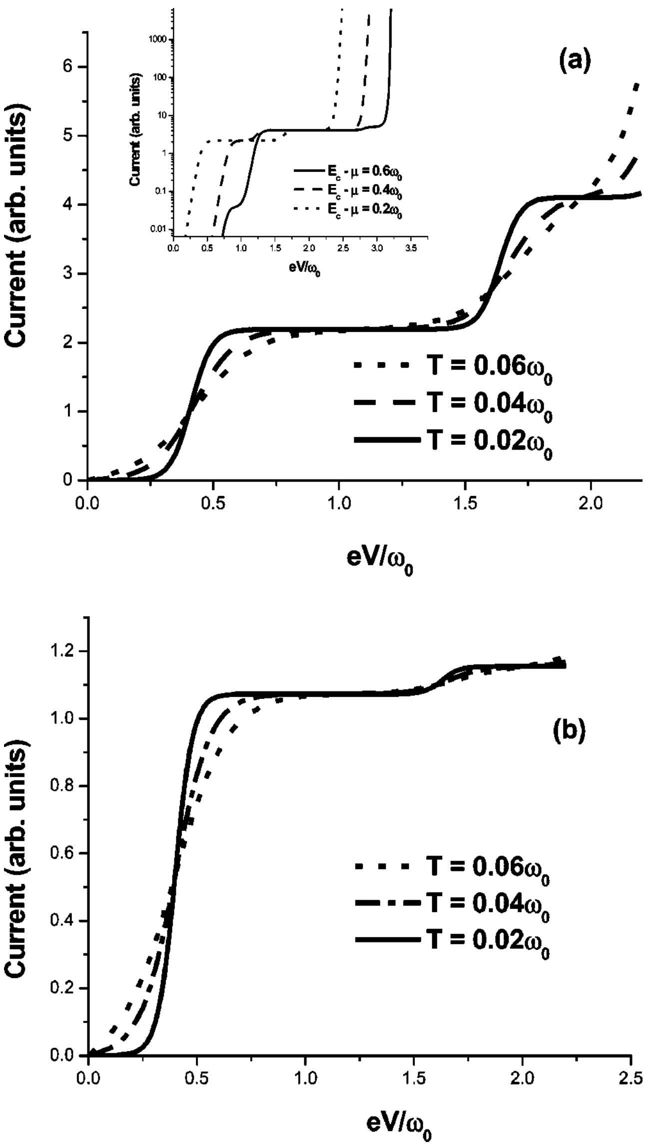

FIG. 1. Current-voltage characteristics of a quantum shuttle system at $E_0-\mu=0.2\omega_0$ and at various temperatures; (a) for the quantum parameter $\nu_0=0.7$, (b) for the quantum parameter $\nu_0=0.07$. Inset: Instability of electron transport at higher voltages for various separations of the electron energy level in the shuttle from the equilibrium chemical potential of the leads.

decrease of current as temperature increases is supplanted by a strong exponential growth for larger values of ν_0. Such an exponential temperature dependence of current-voltage characteristics was also predicted previously for a model of a simple tunnel junction having matrix elements modulated by vibrational motion.[17,21] In this model,[22] the position of the oscillator directly affects the probability of electron tunneling between leads, with integration over the distribution functions of the electrons in the leads subsumed in state densities that are independent of temperature. Correspondingly, the exponential temperature dependence, originating in fluctuations of the oscillator displacement, occurs for all levels of zero-point mechanical fluctuations and, in particular, for $\nu_0 \ll 1$. The present study reveals that the inclusion of a

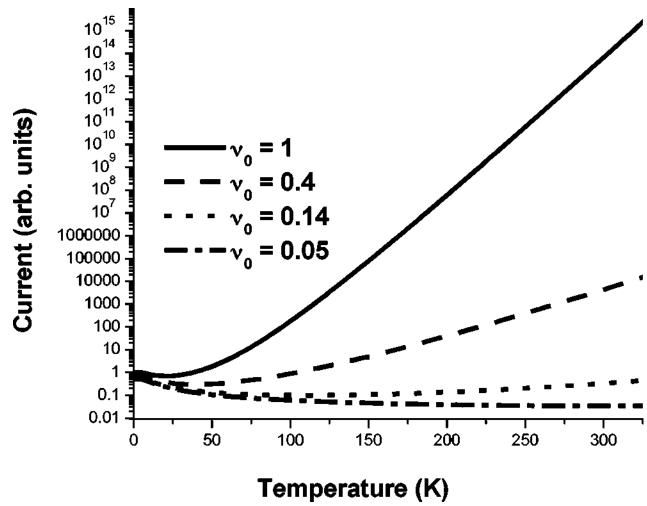

FIG. 2. Temperature dependence of electric current through the shuttle for various values of the quantum parameter ν_0.

single-electron state in the vibrating molecule leads to an additional factor $1/T$ in the formulas for current and conductance (caused by the temperature dependencies of the distribution functions of electrons in the leads). This factor smooths the exponential T dependence in the case of small ν_0. Indeed, for high temperatures ($T \gg |E-\mu_\alpha|$) we find that $\nu_c=\nu_0(2T/\hbar\omega_0)$, so that the current, Eq. (7), is described by the formula

$$I=e\Gamma\,\exp(4T\nu_0/\hbar\omega_0)\frac{eV}{4T}, \tag{16}$$

wherein the temperature dependence involves not only the exponent $e^{4T\nu_0/\hbar\omega_0}$ (as for the tunnel junction coupled to the mechanical oscillator[17,21]) but also involves the factor $1/T$. The competition between these two factors depends crucially on the value of the relative level of zero-point mechanical fluctuations ν_0. At small ν_0 the factor $1/T$ dominates, whereas at larger ν_0 the current-temperature curve demonstrates a very pronounced exponential temperature dependence. However, ν_0 depends on the inverse tunneling length squared, and even small variations in it can bring about a completely different behavior. Accordingly, the experimental data of Refs. 18,19 might correspond to slight differences in connections between molecules and leads leading to very different temperature dependencies of the electron currents. It should be noted that the results of experiments[18] can also be explained by direct electron tunneling.

In conclusion, we have analyzed electron transport through a quantum shuttle having a single-electron energy level. We have shown that at low temperatures there are pronounced steps in the current-voltage characteristics corresponding to direct tunneling through the shuttle and to tunneling accompanied by the absorption and subsequent emission of virtual quanta of mechanical motion (phonons) by the electron. We also found that when the applied voltage facilitates the emission of a real phonon during electron tunneling from the left lead to the shuttle, instability of electron conduction occurs. The temperature dependence of the current has been determined in the range from 2 K to 300 K. Mechanical motion of the shuttle leads to an exponential temperature dependence while the temperature dependence of the electron distribution function in the leads introduces a factor $1/T$ at high temperatures. Competition between these two factors gives rise to a wide variety of current-temperature curves depending on quantum parameter ν_0, such as current decreasing with temperature (at small ν_0), a weak temperature dependence (for intermediate values), and a strong exponential growth (at large ν_0). Such a variety of temperature dependencies was previously demonstrated in experiments on electron transport through long molecules.[18,19]

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