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Effects of the Franck-Condon blockade in tunneling of spin-polarized electrons in a molecular transistor

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We consider a molecular transistor, where the molecule is placed between two fully oppositely spin-polarized electrodes in an external magnetic field. We take into account quantum oscillations of the molecule centrum of mass along the axis, connecting two electrodes. The electric current and conductance are calculated using the equations of motion method and the perturbation theory over the energy level broadening (weak tunneling limit). The Franck-Condon blockade of the current emerges for a strong electron-vibron interaction. However, in our model for the certain values of an external magnetic field, the low-temperature current in the Franck-Condon blockade regime increases. The matter is that the electron energy level in the dot come into voltage transparency "window" depending on the field value. The temperature dependencies of the resonance conductance peaks are also obtained. They show an anomalous (non-monotonic) behavior at intermediate temperatures for a wide range of external magnetic fields in the case of strong electron-vibron coupling. The anomaly occurs due to the interplay of the values of external magnetic field and temperature.

Keywords: single-electron tunneling; molecular transistor; Franck-Condon blockade; spin filters.

Розглянуто молекулярний транзистор, де молекула поміщена між двома повністю поляризованими за спіном електродами, у зовнішньому магнітному полі. Враховуються квантові осциляції центра мас молекули вздовж вісі, що з'єднує електроди. Електричний струм та кондактанс розраховані з використанням методу рівнянь руху та теорії збурень за шириною рівня енергії (слабке тунелювання). При сильній єлектрон-вібронній взаємодії виникає Франк-Кондонівська блокада струму. Однак, в нашій моделі при певних значеннях магнітного поля струм в режимі Франк-Кондонівської блокади збільшується. Причина цього в тому, що електронний рівень на молекулі потрапляє до "вікна" прозорості за напругою залежно від величини зовнішнього поля. Також отримані температурні залежності резонансних піків кондактансу. Вони мають аномальну (немонотонну) поведінку при проміжних температурах у широкому діапазоні зовнішніх магнітних полів при сильній електрон-вібронній взаємодії. Аномалія виникає через накладання ефектів впливу зовнішнього магнітного поля та температури.

Ключові слова: одноелектронне тунелювання; молекулярний транзистор; Франк-Кондонівська блокада; спінові фільтри.

Рассмотрен молекулярный транзистор, где молекула помещена между двумя полностью поляризованными по спину электродами, во внешнем магнитном поле. Учитываются квантовые осцилляции центра масс молекулы вдоль оси, соединяющей электроды. Электрический ток и кондактанс рассчитаны с использованием метода уравнений движения и теории возмущений по уширению уровня энергии (слабое туннелирование). При сильном электрон-вибронном взаимодействии возникает Франк-Кондоновская блокада тока. Однако, в нашей модели при определенных величинах магнитного поля ток в режиме Франк-Кондоновской блокады увеличивается. Причина этого в том, что электронный уровень энергии на молекуле попадает в «окно» прозрачности по напряжению в зависимости от величины поля. Также получены температурные зависимости резонансных пиков кондактанса. Они имеют аномальное (немонотонное) поведение при промежуточных температурах в широком диапазоне внешних магнитных полей при сильном электрон-вибронном взаимодействии. Аномалия возникает из-за наложения эффектов влияния величин внешнего магнитного поля и температуры.

Ключевые слова: одноэлектронное туннелирование; молекулярный транзистор; Франк-Кондоновская блокада; спиновые фильтры.

Introduction

The branch of mesoscopic physics covering the single molecular transistors is a rapidly developing one. The experimental realization of these challenging devices has become feasible since the 2000th [1-3]. Among the novel phenomena, predicted to be observed in these systems, there are the effects based on the vibron-assisted tunneling [4, 5]. The transport properties of tunnel devices change when the electronic states on the electrodes couple to the low-energy vibrational states of the molecule. It is well known [6] that the current through a molecular transistor is a step-like function of the bias voltage with the so-

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called Franck-Condon steps. Each step appears when a new inelastic vibron channel opens due to the extension of an energy "window". For large electron-vibron coupling constants $\lambda \gtrsim 2$ the tunnel current is strongly suppressed. A strong suppression of the current at low bias voltages in molecular transistors is called the Franck-Condon blockade [6] or the "polaronic" blockade.

The vibrational subsystem strongly influences on the resonance conductance and temperature dependence of the conductance peaks. The electron-vibron interaction leads to a rapid decay of the conductance at low temperatures due to the Franck-Condon narrowing of the tunnel broadening of the resonant energy level.

A model of the single-molecular transistor is taken to be used in spintronics. We place the vibrating quantum dot (QD) with a single electronic energy level between two fully spin-polarized electrodes (Fig.1). The magnetization vectors of the electrodes are antiparallel. An external magnetic field perpendicular to the plane of magnetization is applied. For this configuration of the magnetization of the electrodes, the electric current is blocked ("spin blockade" [7]). The external magnetic field induces the spin-flips in QD and lifts the "spin-blockade". The model was suggested in [7] to study the shuttling of the spin-polarized electrons. In [8], the theory of magnetic shuttle was developed by using the same model we use. The influence of a partial spin polarization of the electrodes on the mechanical instability in molecular transistors was studied in [9], [10].

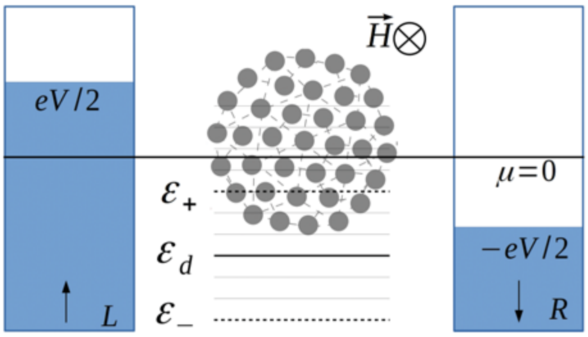


Fig. 1. Model. The vibrating quantum dot containing a single electronic level is placed between two fully spin-polarized electrodes (index $L \equiv \uparrow, R \equiv \downarrow$) with the chemical potentials $\mu_{L,R} = \varepsilon_F \pm eV/2$. The magnetizations of the electrodes are antiparallel (arrows). The direction of the external magnetic field is perpendicular to the direction of spin polarization of the electrodes. Graphic interpretation of levels arrangement is given. The level in the quantum dot is shifted due to the electron-vibron interaction. The Zeeman splitting produces the levels $\varepsilon_{+,-} = \varepsilon_d \pm g\mu_B H/2$.

The goal of the present paper is to study polaronic effects in the tunneling of the spin-polarized electrons through a vibrating quantum dot in an external magnetic field. The average current is calculated using the equations of motion method and the perturbation theory over the level width Γ. The obtained current-voltage characteristics (Fig. 2(a,b)) and temperature dependences of the resonant conductance peaks (Fig. 3(a,b)) illustrate a specific feature of this device to increase the current even for a strong electron-vibron coupling. Furthermore, the temperature dependences of the resonant conductance peaks show an anomalous behavior at intermediate temperatures depending on an external magnetic field.

The Hamiltonian and the equations of motion

The Hamiltonian of a vibrating quantum dot between massive electrodes in an external magnetic field consists of several parts

$$\hat{H}_0 = \hat{H}_l + \hat{H}_{db} + \hat{H}_{int} + \hat{H}_M + \hat{H}_U + \hat{H}_t . \quad (1)$$

The Hamiltonian of electrons in two fully spin-polarized electrodes ($\sigma = L, R = \uparrow, \downarrow$) reads

$$\hat{H}_l = \sum_{k\sigma} \varepsilon_{k\sigma} a^+_{k\sigma} a_{k\sigma} \quad (2)$$

Here $\varepsilon_{k\sigma}$ is the electron energy, $a^+_{k\sigma}(a_{k\sigma})$ is the creation (annihilation) operator for an electron with momentum k and spin projection $\sigma = \uparrow, \downarrow$. The single-level vibrating quantum dot is described by the term

$$\hat{H}_{db} = \hat{H}_d + \hat{H}_b \quad (3)$$

where

$$\hat{H}_d = \sum_{\sigma} \varepsilon_0 c^+_\sigma c_\sigma \quad (4)$$

,

$$\hat{H}_b = \hbar\omega\, b^+ b \quad (5)$$

Here $c^+(c)$ and $b^+(b)$ is the creation (annihilation) fermionic and bosonic operators respectively, ω is the frequency of vibrations. Furthermore, the electron-vibron interaction term

$$\hat{H}_{int} = \sum_{\sigma} \varepsilon_{int} (b^+ + b) c^+_\sigma c_\sigma \quad (6)$$

describes the electrical coupling of electron in the dot to the vibrational mode via the displacement operator and the operator of particles number on the dot. Here ε_{int} is the electron-vibron interaction energy. The term

$$\hat{H}_M = -\frac{g\mu_B H}{2}(c^+_\uparrow c_\downarrow + c^+_\downarrow c_\uparrow) \quad (7)$$

is the Hamiltonian of the electrons in an external magnetic field, where μ_B is the Bohr magneton, g is the gyromagnetic ratio. The Coulomb interaction term reads

$$\hat{H}_U = U c^+_\uparrow c_\uparrow c^+_\downarrow c_\downarrow , \quad (8)$$

where U is the Coulomb repulsion energy.

The tunneling Hamiltonian describes the electron tunneling between the electrodes and a quantum dot

$$\hat{H}_t = \sum_{k\sigma} t_\sigma a^+_{k\sigma} c_\sigma + h.c. \qquad (9)$$

Here $c^+_\sigma (c_\sigma)$ is the creation (annihilation) operator for an electron in the dot with spin projection σ, t_σ is the tunneling amplitude, which in general case depends on the displacement of the molecule. In what follows, we will assume the tunneling amplitudes to be coordinate-independent. This assumption is verified for the case of strong electron-vibron coupling (see, e.g., [11]).

Hamiltonian (1) is diagonalized in two steps. Rotation at the angle $\varphi = \pi/4$ leads to the elimination of the term which is non-diagonal on spin projection. New variables are the linear combinations of the dot operators $d_s = (c_\uparrow + j_s c_\downarrow)/\sqrt{2}$, ($s = 1,2;\ j_{1,2} = -1,+1$). The transformation provides the Zeeman splitting of the level in the dot.

Next we apply an unitary transformation with the operator $\hat{V} = \exp(i\sum_{s=1,2} \lambda n_s \hat{p})$, where $\hat{p} = i(b^+ - b)/\sqrt{2}$ and $n_s = d^+_s d_s$ (the so-called Lang-Firsov or "small polaron" transformation [12]). The dimensionless parameter $\lambda = -\sqrt{2}\varepsilon_{int}/\hbar\omega$ characterizes the strength of the electron-vibron coupling. This results in the "polaronic" shift of the electron energy in the dot, which becomes $\varepsilon_d = \varepsilon_0 - \lambda^2\hbar\omega$, in the replacement of the tunneling amplitude by $t_{\sigma s}(\hat{p}) = t_{\sigma s} e^{-i\lambda\hat{p}}$, where $t_{\sigma s}$ is the matrix elements of the matrix

$$\hat{T} = \frac{1}{2} t_\sigma \begin{pmatrix} +1 & +1 \\ -1 & +1 \end{pmatrix} \qquad (10)$$

and in the shift of electron-electron correlation energy $\tilde{U} = U - \lambda^2\hbar\omega$. In what follows we assume that $\tilde{U}$ is small and neglect the effects of electron-electron correlations.

The transformed Hamiltonian reads

$$\hat{H} = \hat{H}_l + \hat{H}_b + \hat{H}'_d + \hat{H}'_t, \qquad (11)$$

where

$$\hat{H}'_d = \sum_s (\varepsilon_d - \tfrac{1}{2} j_s g\mu_B H) d^+_s d_s, \qquad (12)$$

$$\hat{H}'_t = \sum_{k\sigma s} t_{\sigma s} e^{-i\lambda\hat{p}} a^+_{k\sigma} d_s + h.c. \qquad (13)$$

In order to calculate the current through the system, we find the time dependences of the fermionic and bosonic operators using Heisenberg approach. In a wide-band approximation the time dependences of fermionic operators become the functions of the partial energy level widths $\Gamma_{\sigma s} = 2\pi\nu(\varepsilon_F) t^2_{\sigma s}$. For a weak tunneling, we assume $\Gamma_{\sigma s}$ to be the smallest energy parameters of the problem, which, however, cannot be neglected for the equations of motion for fermionic operators. On the other hand, in the perturbation approach with respect to $\Gamma_{\sigma s}$ the equation for bosonic operator is reduced to a harmonic oscillator equation.

We consider a case of the symmetric tunnel junction ($\Gamma_L = \Gamma_R \equiv \Gamma$), thus the set of bound equations of motion for fermionic operators on the dot is readily decomposed to two independent differential equations.

The average electric current and the conductance

The operator of the electric current in the $\sigma(L,R)$-electrode reads

$$\hat{I}_\sigma = -e\frac{d\hat{N}_\sigma}{dt}, \qquad (14)$$

where $\hat{N}_\sigma = \sum_k a^+_{k\sigma} a_{k\sigma}$ are the particle number operators. Then the average current can be expressed as

$$I_\sigma = -\frac{2e}{\hbar}\sum_s t_{\sigma s}\,\mathrm{Im}\left\langle \sum_k a^+_{k\sigma}(t) d_{s\,p}(t) \right\rangle_{\hat{H}}, \qquad (15)$$

where the average is taken with respect to the full Hamiltonian. In perturbation theory, the average can be factorized into the product of two averages, each depending only on one type of quasiparticles − fermions or bosons. The averages then can be taken with the unperturbed fermionic Hamiltonian and unperturbed bosonic Hamiltonian, respectively. Following the calculation procedure of [13], we find that the steady state current can be written as a sum of partial currents over "vibron channels"

$$I = I_{\max} \sum_{n=-\infty}^{+\infty} A_n(\omega) \sum_{s=1,2} \{ f_L(\varepsilon_s) - f_R(\varepsilon_s) \}. \qquad (16)$$

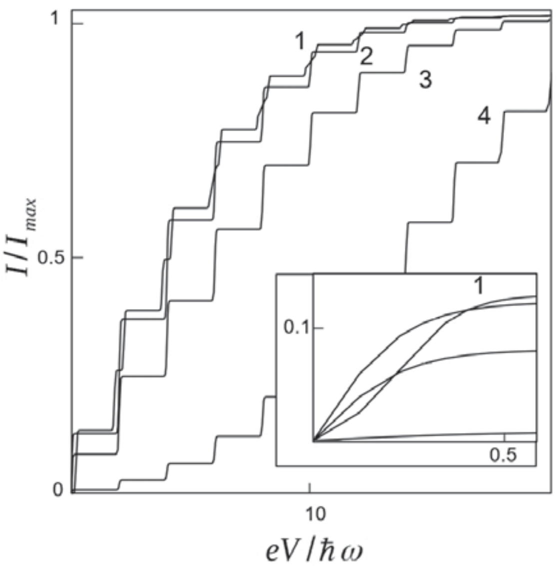
Here $\varepsilon_s = \varepsilon_d - n\hbar\omega + j_s\tilde{H}$, $\tilde{H} = g\mu_B H/2$, and

$$I_{\max} = \frac{e\Gamma}{4\hbar}\frac{\tilde{H}^2}{\tilde{H}^2 + (\Gamma/2)^2} \qquad (17)$$

is the maximal (saturation) current, $f_\sigma(\varepsilon) = (\exp[(\varepsilon - \mu_\sigma)/k_B T] + 1)^{-1}$ are the Fermi-Dirac distribution functions. The spectral weights $A_n(\omega)$ are

$$A_n(\omega) = e^{-\lambda^2(1+2n_B)} I_n(z) e^{-\frac{\hbar\omega}{2k_B T}n}, \qquad (18)$$

where $I_n(z)$ denote the modified Bessel functions, $z = 2\lambda^2\sqrt{n_B(1+n_B)}$, $n_B = (\exp[\hbar\omega/k_B T] - 1)^{-1}$ is the Bose-Einstein distribution function.



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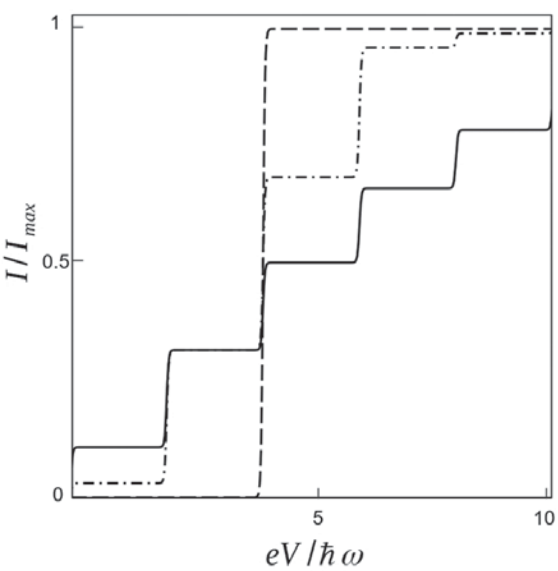
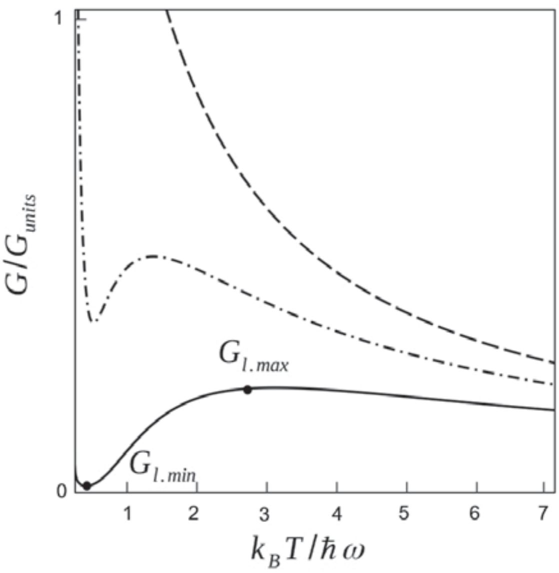


b

Fig.2. Current-voltage characteristics of the molecular transistor with spin-polarized electrodes in an external magnetic field. 2(a): Current-voltage characteristics for the fixed coupling constant $\lambda = 3$ and different values of an external magnetic field: $\tilde{H} / \hbar\omega$ =0.1(curve 1), 1 (curve 2), 3 (curve 3), 7 (curve 4). The inset shows the same dependences in the range of low biases. 2(b): Current-voltage characteristics for a fixed value of the external field $\tilde{H} / \hbar\omega$=2 for different electron-vibron coupling constants: $\lambda = 0$ (dashed line), $\lambda = 2$ (dash-dotted line), $\lambda = 3$ (solid line). $\Gamma / \hbar\omega = 0.001$, $k_B T / \hbar\omega = 0.01$ for all calculations. We set $\mu_\sigma |_{V=0} = \varepsilon_F$ and $\varepsilon_0(V_g) - \varepsilon_F = 0$, where V_g is the gate voltage, thus $\mu = 0$ coincides with ε_0.



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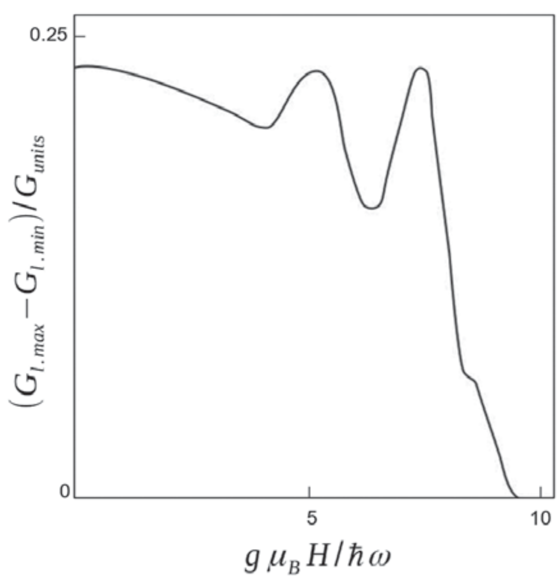

b

Fig.3. Anomalies in temperature dependence of the conductance resonance peaks. 3(a): The temperature dependence $G(k_B T / \hbar\omega)$ in units $G_{units} = (k_B T / \hbar\omega) G_{\tilde{H}}(T)$ at a fixed value of the external magnetic field $\tilde{H} / \hbar\omega = 1$ for different coupling constants: $\lambda = 1$ (dashed line), $\lambda = 2$ (dash-dotted line), $\lambda = 3$ (solid line). $\Gamma / \hbar\omega = 0.001$. Nonzero coupling causes an anomalous behavior at intermediate temperatures. 3(b): The dependence of $(G_{l.max} - G_{l.min}) / (G_{\tilde{H}} \Gamma / \hbar\omega)$ from $g \mu_B H / \hbar\omega$ for $\lambda = 3$, which describes the presence of the anomaly in a wide range of fields and its non-monotonic behavior when the field increases. We set $\mu_\sigma |_{V=0} = \varepsilon_F$ and $\varepsilon_0(V_g) - \varepsilon_F - \lambda^2 \hbar\omega = 0$, where V_g is the gate voltage. Thus $\mu = 0$ coincides with $\varepsilon_0 - \lambda^2 \hbar\omega$ for each curve.

Differential conductance in the linear response regime is defined as $G = \left. \frac{dI}{dV} \right|_{V \to 0}$. From Eq. (16), one readily gets

$$G = G_{\tilde{H}}(T) \sum_{n=-\infty}^{\infty} A_n(\omega) \sum_{s=1,2} \cosh^{-2}\left(\frac{\varepsilon_s}{2k_B T} \right), \quad (19)$$

with

$$G_{\tilde{H}}(T) = \frac{\pi e^2}{2h} \frac{\Gamma}{k_B T} \chi(\tilde{H}) \quad (20)$$

where $\chi(\tilde{H}) = \tilde{H}^2 / (\tilde{H}^2 + (\Gamma/2)^2)$. Notice that the current and the conductance equal zero at $\tilde{H} = 0$, showing the "spin blockade" phenomenon in our system.

The obtained current-voltage characteristics are plotted in Fig. 2(a,b), where the expected appearance of the Franck-Condon steps of the current due to non-zero electron-vibron coupling is observed. The current-voltage characteristics for different values of an external magnetic field and for fixed coupling constant $\lambda = 3$ at low temperatures are shown in Fig. 2(a). The behavior of the curves strongly depends on the value of an external magnetic field because it controls the appearance of quantum dot energy level in the bias voltage "window", $[\varepsilon_F + eV/2,\ \varepsilon_F - eV/2]$. The fermionic level $\varepsilon_0(\mathrm{V}_g) = \varepsilon_F$ in the dot is shifted on $\Delta_\varepsilon = -\lambda^2 \hbar\omega$ due to electron-vibron coupling and does not fall into transparency "window" at low biases $eV < 2\lambda^2 \hbar\omega$. By changing the external magnetic field one can move the upper Zeeman split level ($\varepsilon_+ = -\lambda^2 \hbar\omega + \tilde{H}$) to this window and therefore strongly enhance the current at finite temperatures. Notice that the Franck-Condon blockade of the current (current is proportional to $e^{-\lambda^2}$) at low temperatures $k_B T << \hbar\omega$ and low biases ($eV << \hbar\omega$) formally originates from the summation in Eqs. (16), (18) over non-positive n ($n = 0, -1, \ldots$). By adjusting $\tilde{H}$, one can move dot level to the transparency "window" and therefore increase the current. Thus the current can be controlled in the same way as it could be controlled by using the gate electrode. However, the Franck-Condon blockade cannot be lifted by the external magnetic field as the current is still suppressed comparing to the low-bias current in the absence of electron-vibron interaction.

Fig. 2(b) shows $I - V$ characteristics at a fixed field $\tilde{H} = \hbar\omega$ for different coupling constants λ. The curves illustrate that the device supports the low bias current, while the Zeeman splitting of levels blocks it in the absence of electron-vibron interaction when $eV < \tilde{H}$. Moreover, the current increases for an appropriate field values $\tilde{H}$, when the electron-vibron coupling constant λ increases. Usually, for large coupling constants λ, the current is suppressed in SMT when the magnetic field is not applied.

Now we pass to the study of the temperature dependence of the resonance peak of differential conductance at $V = 0$. The curves $G(k_B T / \hbar\omega)$ are shown in Fig.3 (a) for a different coupling constant λ at the fixed field value $\tilde{H} = \hbar\omega$. The asymptotic expressions for low- and high-temperature conductance read

$$\frac{G(T)}{G_{\tilde{H}}(T)} = \begin{cases} e^{-\lambda^2}, & \Gamma << k_B T << \hbar\omega, \\ 1 - \frac{\lambda^2 \hbar\omega}{2k_B T}, & k_B T >> \lambda^2 \hbar\omega. \end{cases} \quad (21)$$

At the low temperatures, $k_B T << \hbar\omega$, the conductance is suppressed due to the strong electron-vibron coupling. At the high temperatures, $k_B T >> \lambda^2 \hbar\omega$, polaronic effects vanish, and the conductance has the same behavior as that for a quantum dot (in the first term of the relation obtained in this limit), because all inelastic channels are open and the Franck-Condon blockade is lifted. At intermediate temperatures, $k_B T \gtrsim \hbar\omega$, for strong electron-vibron interaction the dependences are highly non-monotonic due to the interplay between the temperature and external field effects. An anomalous behavior is observed for the curves at different values of the field.

In order to qualitatively describe this anomaly, we introduce the local maximum $G_{l.max}$ and minimum $G_{l.min}$ of the conductance in this temperature range (see Fig.3(a)). The value $G_{l.max} - G_{l.min}$ is a non-monotonic function of the external magnetic field $\tilde{H}$, as shown in Fig.3 (b) for $\lambda = 3$. For large values of the external magnetic field $\tilde{H} >> \lambda^2 \hbar\omega$, the difference $G_{l.max} - G_{l.min}$ tends to zero, and the anomaly vanishes.

Conclusions

The current in the single molecule transistor with fully oppositely spin-polarized electrodes in an external magnetic field is evaluated. The limit of weak tunnel coupling $\Gamma / k_B T << 1$ is considered. The current-voltage characteristics demonstrate the Franck-Condon blockade for strong electron-vibron coupling and show the standard Franck-Condon steps. We show that in this model the external magnetic field allows to control and increase low-temperature current. It is also shown that the electron-vibron strongly affects the transport properties of the system. The current rises (in comparison with the current through a non-movable quantum dot) at the voltages $eV < \tilde{H}$ regardless of the Zeeman splitting of energy level due to the vibron-assisted tunneling. In addition, the low-bias current may increase at the certain conditions, when the electron-vibron interaction increases.

The dependence of the resonance conductance peaks on the temperature is highly non-monotonic. This anomalous dependence takes place at intermediate temperatures $k_B T \gtrsim \hbar\omega$, where there is a transition from the regime of the Franck-Condon blockade at low-temperatures to the regime of $1/T$ -decay at high temperatures. In this region difference between the local maximum and local minimum of the conductance depends on the magnetic field in a non-monotonic way. The obtained dependences can be used to control the current and conductance in the magnetically operated molecular transistors.

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