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Modelling of Cold Electron Filtration in Tunnelling Nanostructures with Metallic Quantum Dot

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The microscopic model of suppressing thermal smearing of electrons in resonant tunnelling structures with nonlinearity due to electron-phonon interaction under the assumption of the presence of degenerate levels in the quantum well is proposed. The electron-phonon interaction results in an effective attraction with breaking the degeneracy and lowering the energy level. We note the manifestation in this model of the Fano effect associated with multichannel tunnelling. As an example, the effect of suppressing the thermal spread of electrons in a nanotransistor with a metal quantum dot is considered and compared with the experiment. A rather significant (about 200 K) heating of the quantum dot was detected caused by the passage of a current through the nanotransistor. Despite heating, the effect of cold electrons filtration is clearly observed. The theory and experiment were found to be in a good agreement.

Key words: resonant tunnelling, quantum dot, electron-phonon interaction, degenerate levels, filtration of electron thermal smearing.

Запропоновано мікроскопічну модель пригнічування температурного розподілу електронів у резонансних тунельних структурах з нелінійністю, обумовленою електрон-фононою взаємодією в припущенні наявності вироджених рівнів у квантовій ямі. Електрон-фононна взаємодія призводить до ефективного притягання зі зняттям виродження та зниженням енергетичного рівня. Ми відзначаємо, що у запропонованій моделі проявляється ефект Фано, пов'язаний з багатоканальним тунелюванням. Як приклад, розглянуто ефект пригнічення теплового розкиду електронів в

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нанотранзисторі з металевою квантовою точкою. Виявлено досить значне (близько 200 K) нагрівання квантової точки, викликане проходженням струму через нанотранзистор. Незважаючи на нагрівання, ефект фільтрації холодних електронів чітко спостерігається. Теорія та експеримент добре узгоджуються.

Ключові слова: резонансне тунелювання, квантова точка, електрон-фононна взаємодія, вироджені рівні, фільтрування термічного розкиду електронів.

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1. INTRODUCTION

The use of nanodevices based on quantum dots for information processing seems to be very promising for the practical application [1, 2]. However, their characteristics are washed out at room temperatures, which is a serious drawback to a practical use. It seems very actual to overcome this objection.

Recently, several groups of researches [3, 4] reported about a few nanometre silicon point-contact quantum dot (QD) (the so-called ‘single-electron transistor’), which can operate at room temperature. It was shown that if the QD size is sufficiently small (<10 nm), any spacing between excited states is much greater than $kT \approx 25$ meV such that these states control electron tunnelling across the QD even at room temperatures.

In Ref. [5] it was reported about a nanotransistor with the property of controlled changing of conductivity that was not exposed to temperature. The device consists of a nanoparticle of a size of 10 nm made of a semiconductor or of a metal (served as a QD) embedded in a matrix of SiO_2 that was positioned on an insulating layer of Cr_2O_3 .

The authors of this paper explained the phenomenon by taking into account two factors. The first one is an existence of the Coulomb blockade, which leads to the appearance of current steps in current-voltage characteristics. The second one is the formation of a three-barrier structure, in which current carriers pass sequentially through the quantum well and QD. In such a structure, the suppression of the thermal spread looks like a successive transition of electrons between levels with emission of phonons [6]. However, the emission of phonons automatically leads to heating of the nanostructure, which was not taken into account in this work.

In contrast to Ref. [5], in our previous paper [7], we have shown that the appearance of the conductive peaks is due to the spatial quantization in a nanoscaled semiconductor QD. The states in the QD are degenerate and can be split as a result of the electron-phonon interaction. The splitting energy levels can have lower energies than the Fermi en-

ergy of the source electrode. Such an approach permits to describe experimentally observed suppressing of temperature smearing in the tunnelling device.

In the present paper the thermal smearing suppression concept proposed in our early paper [7] is used to explain the filtration ability of a tunnelling device with a metal nanoparticle as a QD that was experimentally studied in Ref. [5]. In the experiments with a metal nanoparticle, the gate electrode was added to the device.

2. ENERGY SPECTRUM OF A METAL NANOPARTICLE

Unique properties of nanoobjects are due to a spatial quantization leading to an essential transformation of their spectrum. A metal nanoparticle (~10 nm Au nanoparticle) embedded in a dielectric matrix and connected through tunnelling barriers to two electrodes is called a 'quantum dot (QD). A dot with a linear size L forms a one-dimensional box with energy levels

$$E(v) = \frac{\pi^2 \hbar^2 v_L^2}{2m_e L^2} + E_{\min}, \quad (1)$$

where $v_L = 0, \pm 1, \pm 2, \dots$ and m_e is an electron mass. Its spectrum is given in Fig. 1. Since the conduction band of the metal QD is completely occupied by electrons, then $E_{\min} = \min E_v$, where E_v is the electron energy level in the conduction band. In the following we set $E_{\min} = 0$.

It can be seen in Fig. 1 that the energy spectrum of the nanoparticle acquires a discrete character. The allowed state with a quantum number of v_μ corresponds to the energy $E(v_\mu) = E_\mu$. Using Eq. (1) we get $v_\mu = L\sqrt{2m_e E_\mu} / \pi\hbar$. For nanoparticles made of gold, the Fermi energy equals $E_\mu = 5.54$ eV. The allowed states are formed due to a spatial quantization in a cavity having the particle size, hence the electron mass in the QD will correspond to the mass of a free electron. Then for the μ -electron state in the QD with $L = 10$ nm, the quantum number will be of $v_\mu \approx 40$.

In the case of the spatial quantization, the distance between energy levels is not a constant value. It depends on the quantum number v_μ as follows from Eq. (1)

$$\begin{aligned} \Delta E_1 &= E(v_\mu - 1) - E(v_\mu) = \varepsilon_0 (2v_\mu - 1), \\ \Delta E_2 &= E(v_\mu + 1) - E(v_\mu) = \varepsilon_0 (2v_\mu + 1). \end{aligned} \quad (2)$$

Here $\varepsilon_0 = \pi^2 \hbar^2 / 2m_e L^2$.

Experimental plot [5] of the current J through the tunnelling device as a function of the gate voltage V_g is shown in Fig. 2. Due to large current fluctuations observed in the plot, a small change in the spacing

between the levels is difficult to identify. To define a number ν_μ from the experimental data, it is appropriate to use the following relation $\Delta E = (\Delta E_2 + \Delta E_1) / 2 = 2\varepsilon_0 \nu_\mu$. From the experimental plot in Fig. 2, we get the value $\Delta E \approx 0,3$ meV. Taking into an account that $\varepsilon_0 \nu_\mu^2 = 5.54$ meV, the following values can be found $\varepsilon_0 \approx 4.05$ meV and $\nu_\mu = 37$. These values are close to those that were calculated with the use of the expression (1).

From these considerations we can conclude that the experimentally observed peaks in the current dependence on the gate voltage are most likely due to a spatial quantization.

3. THEORETICAL MODEL

It follows from Fig. 1 that the allowed states in a QD are degenerate by a quantum number n and spin. Thus, every quantum level may be at least four-fold degenerate. If electron-electron or electron-phonon interaction is taken into account, the degeneracy may be removed.

We calculate the tunnelling current through the allowed four-fold degenerate level with the use of the results of our previous papers [7–10]. Here we consider only situations where the applied voltage between the two electrodes is small, so the system is in a steady state.

Fig. 1. Energy spectrum of a metal nanoparticle embedded in a dielectric matrix. μ_L , μ_R , μ are chemical potentials in left, right electrodes, and a nanoparticle, respectively. Dashed lines correspond to the allowed electron states and symbol • denotes the states occupied by electrons.

The current through a QD having the lowest energy $E(v, m) \equiv E_{vm}$, where $m = 0, 1, 2, 3$, will be determined by the relation

$$J_{cd} = \sum_v J_{cd}(v). \quad (3)$$

The interaction between electrons lifts the degeneracy and the energy E_m will depend on a number m . In the case when $\Delta E_v \gg k_B T$, where k_B is a Boltzmann constant and T is temperature, $J_{cd}(v)$ can be calculated independently for each level v by the formula [7–10]:

$$J_{cd}(v) = \sum_{m=0}^3 J_0(E_{vm}) \{f_L(E_{vm}) - f_R(E_{vm})\} (1-n)^{3-m} n^m C_3^m + J_s, \quad (4)$$

where $J_0(E_{vm}) = \frac{e}{\hbar} \frac{\Gamma_R(E_{vm})\Gamma_L(E_{vm})}{\Gamma_R(E_{vm}) + \Gamma_L(E_{vm})}$ and n is the average value of the occupation number of a splitting energy level; $\Gamma_L(E_{vm})/\hbar$ and $(\Gamma_R(E_{vm})/\hbar)$ are the rates of electron passing through left and right barriers, respectively; $f_{L,R}(E_{vm}) = [\exp((E_{vm} - \mu_{L,R})/k_B T) + 1]^{-1}$ are Fermi–Dirac distribution functions in leads, the chemical potentials in the left and right leads, $\mu_{L(R)}$, respectively, are related to the bias V and gate V_g voltages; $E_{vm} = E_v + V_g + mU - \gamma V$ are the energies of the allowed states in a QD.

In the case of a metallic particle, $E(v_\mu) = \mu$ and $\gamma = \alpha_L / (\alpha_L + \alpha_R)$, where α_L and α_R are the widths of the left and right barriers, respectively. In the experimental conditions $\alpha_R \approx \alpha_L$ so, $\gamma = 0.5$. The chemical potentials of the left and right leads are connected by the relation

Fig. 2. Demonstration of the experimentally observed [5] differences in energy spacing $v_\mu - (v_\mu - 1)$ and $(v_\mu + 1) - v_\mu$ ($\Delta E_2 > \Delta E_1$) on the plot of the current versus the gate voltage.

$\mu_L = \mu_R + V$. In the following we set $\mu = \mu_L$. The parameter $U = U_c - U_{ph}$ defines the value of energy splitting. It consists of the two terms, one is a Coulomb repulsion U_c and the other, U_{ph} , is due to the interaction between electrons through phonons leading to the attraction of electrons. The current J_s is due to the transmission of electrons through other channels and it is rather small to be neglected.

In the expression for the current (4) the main parameter is the occupation n of the splitting levels after the removal of the degeneracy. This number as is shown in Refs. [7–10] can be calculated using the equation

$$n = F(n), \quad (5)$$

where

$$F(n) = \sum_{m=0}^d C_d^m g_m (1-n)^{3-m} n^m, \quad C_d^d = \frac{d!}{m!(d-m)!}, \quad (6)$$

$$g_m = g(E_m), \quad g(E) = \frac{\Gamma_L(E)f_L(E) + \Gamma_R(E)f_R(E)}{\Gamma_L(E) + \Gamma_R(E)}.$$

In the following, we'll use the assumption that $\Gamma_L(E_m) = \Gamma_R(E_m) = \Gamma$ are constant values.

4. DEPENDENCE OF LEVEL OCCUPATION NUMBERS ON THE GATE VOLTAGE

In this Chapter we'll consider a level occupation of a four-fold degenerate state when its position shifts with respect to the chemical potential of electrodes. This shift can be caused by the gate voltage V_g which controllably tunes the energy levels in the QD. When the value of V_g is such that $E_m > \mu$ for all m numbers ($m = 0, 1, 2, 3$), then the QD is not filled with electrons and the degeneracy is not removed, as demonstrated in Fig. 3, *a*. With increasing of the gate voltage, the value of E_1 reaches the value of the chemical potential ($E_1 = \mu_L$, but $E_0 > \mu_L$). At that moment, the energy level splits abruptly (see Fig. 3, *b*).

Every splitting level will be occupied by electrons. However, an electron on the level E_0 may have the energy that exceeds the Fermi energy on the value U . This follows from the fact that the total electron energy

$$W = \sum_{m=0}^{d-1} (E_m - \mu) \text{ will be negative in this case.}$$

In the range of voltage values at which $E_1 < \mu < E_2$ in accordance with Eq. (6), the system becomes bistable and may be in three states. The states with occupation numbers $n_1 = 0$ and $n_2 = 1$ are stable, and

the state with $n_2 = 0.5$ is unstable. The appearance of bistability is due to the requirement of a stepwise change in the energy of electrons in the QD by U . Therefore, the QD with a negative energy of splitting can save the state of electrons created at the previous value of V_g . Here, an analogy is observed with the effect of a state memory, which was described in Refs. [10–12].

For the numerical simulation of the current J versus V_g dependences we take the value of the bias voltage V to be 10 meV as in the experiment of the Ref. [5].

When the gate voltage V_g is such that $E_0 = \mu$, then $f_L(E_m) = f_R(E_m) = 1$. The degeneracy will be removed and all levels in the QD will be filled by electrons. As V_g is increased, the distribution functions will take the values close to zero. But electrons that have occupied levels E_0, E_1 will block the transport through them as is shown in Fig. 4, *a*. This situation will be continued until V_g reaches a value at which $E_1 = \mu - V_g$.

When $E_1 > \mu - V_g$, the occupation of states changes dramatically becoming partial, and tunnelling through these states is carried out as shown in Fig. 4, *b*. The current sharply increases, while the electrons participating in the process are injected with an energy lower than the Fermi energy. With a further increase in the gate voltage, when the value $E_3 \geq \mu - V_g$ is reached, the current decreases sharply, as shown in Fig. 4, *c*. The dependence of J vs V_g has a bell-shaped character with a maximum at the voltage $V_{gm} = \mu + 3U/2$. Then with the increase of V_g , the occupation of the QD drops to zero.

The dependence of level occupancies in the QD on the gate voltage shown in Fig. 4, *b* illustrates the role of the degenerate level splitting in suppressing thermal spread of current carriers.

Fig. 3. Dependence of the QD occupation on the mutual position of energy levels with respect to the chemical potential of electrodes. The gate voltage shifts energy levels of the QD by the value of V_g with respect to chemical potential μ . The case when $E_1 > \mu$ (*a*). Breaking of degeneracy at $E_1 < \mu$ (*b*). Hysteresis in the dependence of the QD occupation n on V_g (*c*).

The calculated dependencies of the current J and occupation n of levels on the gate voltage V_g at different values of level splitting U with the value of the bias V equal to 10 meV is shown in Fig. 5.

It can be seen that with increasing of $|U|$ (U being negative), the current peak narrows and its maximum shifts by the value $-3|U|/2$ and at the same time the peak narrows. Narrowing is due to an increase in the rate of change in the filling of quantum levels from zero to unity. In general, the width of the current peak is determined by the voltage range in which the population of quantum levels varies from zero to unity (see Fig. 5, *b*). At sufficiently large values of $|U|$ (comparable with $k_B T$), the transition occurs very quickly and is practically independent of temperature.

In an experiment, voltage fluctuations can play a significant role. In the interval where the sharp increase of the level occupancy takes place, small changes in the voltage cause large changes of the current as is shown in Fig. 5 (curve 3) with a negative value of U . For the simulation of fluctuations, the equation $\Delta V_g = r_0(1 - 2r)$ was used where r_0 is the amplitude of fluctuations and r ($0 \leq r \leq 1$) takes random values. The obtained dependence (Fig. 6) resembles the typical experimental behaviour [5]. Such a dependence is dictated by a sharp change in the occupation of quantum states from zero to unity, which is a manifestation of the collectivization of electrons in quantum dots. Electrons simultaneously collectively leave the QD or occupy it. Under such conditions, small voltage fluctuations can dramatically change the occupation of the QD.

Figure 7 shows the total current through all resonance levels for $U = -10$ meV and $k_B T = 10$ meV (solid curve). Due to the splitting of the

Fig. 4. Dependence of level occupancies in the QD (*a*, *b*) and current through it (*c*) on the gate voltage V_g . $V_g = 0$ (*a*), current is zero (*b*). V_g shifts splitting levels. Current runs through some of them (*c*).

degenerate level, the maximum of the total current forms at the value of $-3U/2$ with respect to its position without interactions.

5. COMPARISON WITH EXPERIMENT

With the use of Eq. (4) we can calculate the current through the system as a function of the gate voltage using experimental parameters and compare theoretical and experimental dependencies. We'll consider only three levels that are formed in the QD by a spatial quantization and are situated near the Fermi level of the left lead. In Fig. 8, we present the experimental dependences from Ref. [5] and theoretical simu-

Fig. 5. Dependence of the current J (a) and occupation n (b) on the gate voltage V_g at different values of the splitting potential U : $U = 0$; $U = -10$ meV; $U = -20$ meV. Other parameters are the same for all curves and equal to the following values: $V = 10$ meV, $E_0 = \mu = 0$, $J_0 = 140$ pA, $kT = 10$ meV.

lations for different temperatures. System parameters in all cases were the same.

From the experimental plot in Fig. 2, which is also the lowest plot in Fig. 8, the value U can be found from the condition $V_{gm} = -3U/2$ that defines the shift of the peak maximum relative to the position of the level when interactions are absent. Its value appeared to be equal to $U \approx -17$ meV. At the same time from the experimental plots, we can see that $U = -25$ meV what is a sufficiently good agreement. The intervals between peaks appeared to be $\Delta E_1 = 297$ meV and $\Delta E_2 = 303$ meV in all plots.

It was unexpected to us that the result of comparison the theory and experiment was satisfactory only if the heating of the QD when a current flowing through it was taken into account. In this case, the heat-

Fig. 6. Current J and occupancy n dependence on the gate voltage V_g in the case of voltage fluctuations. Parameters of the calculation are: $U = -20$ meV, $J_0 = 140$ pA, $E_0 = \mu = 0$, $kT = 20$ meV, $V = 10$ meV. The solid line corresponds to the absence of fluctuations. Circles connected with thin lines were obtained with the amplitude of fluctuations of the value of $r_0 = 5$.

ing temperature has to be equal to $k_B T_h = 22$ meV. However, this result is predictable, since an estimate of the density of the current will be (at a peak maximum of ~ 10 pA) of approximately ~ 100 A/mm². At such a current density, heating of the QD is natural. The heating phenomenon of QDs, which were fullerene molecules, was also revealed in Ref. [13] by comparing the experimental and theoretical dependences.

By adjusting the theory and experiment, the value J_0 turned out to be equal to 140 meV and was the same for all temperatures. Using the numerical value found for J_0 , the transparency of the tunnel barriers between which the nanoparticle is enclosed can be estimated. It follows from the expression $J_0 = e\Gamma / \hbar = 140$ pA, that $\Gamma \approx 1$ meV.

On the other hand, time that takes an electron to pass through the QD, is $\tau = \hbar / \Gamma$ and according to [14] it corresponds to the value $\tau = L / Q v_F$, where Q is the transparency of the barrier, and v_F is the electron velocity at the Fermi level. For gold it has the value of $v_F = 1.4 \cdot 10^8$ cm/s. Thus, we obtain the value of the transparency of tunnel barriers to be $Q \approx 10^{-2}$. The low value of the transparency justifies the approximations we use.

From Fig. 8 it can be seen that the peaks remain well resolved up to a high temperature.

Fig. 7. The formation of the total current passing through the QD by electrons (solid curve), which are transported through the resonance levels E_0 , E_1 , E_2 , E_3 (thin curves).

6. CONCLUSION

In conclusion, we have analyzed the experiments of Ref. [5], where the suppression of thermal smearing of current carriers in tunnelling systems with a gold nanoparticle as a QD was observed, in terms of a microscopic tunnelling model with account of electron-phonon interactions.

In the behaviour of the system, the manifestation of the Fano effect [15, 16] is clearly seen but with one essential addition—the energy po-

Fig. 8. $I-V_g$ characteristics for different ambient temperatures. Above 10 K, each $I-V_g$ plot is vertically displaced by $m \cdot 15$ Pa for clarity. Solid lines represent simulation results. Circles connected by the thin line are experimental data of Ref. [5]. The bias voltage V was 10 mV.

sition of tunnelling channels depends on an occupancy of the QD by electrons. In each channel, the current-voltage dependence may have an asymmetrical form but the total current through all the channels will be a symmetrical one. In other words, the inversion of the Fano effect is possible, when the symmetrical dependence is formed from asymmetric contributions from each of the channels.

As compared to a semiconductor QD, in a metallic QD a separation between levels is 20 times less and is comparable to a thermal spread of electrons at room temperature. So, at current-voltage dependencies, the steps are significantly smoothed, nevertheless, this turned out to be enough to observe the effect of thermal suppression in the dependences of the current on the gate voltage.

From the comparison of the simulated current-voltage characteristics and the experimental plots, the value of U is confirmed to be negative and significantly large. The question remains, how it can be achieved in such device? The value of U is negative, when the electron-phonon interaction exceeds the Coulomb repulsion. The magnitude of the electron-phonon interaction is inversely proportional to the frequencies of phonons involved in the interaction. It is known that low-frequency vibrations take place on the surface of the nanoparticle. For a nanoparticle of a very small size, surface effects can play a significant role.

We can expect that the device presented in Ref. [5] and analyzed by us in this paper, with a metal QD of a few nanometre sizes, will find practical application in quantum devices at elevated temperatures.

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