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Effects of electron-phonon interaction in tunneling processes in nanostructures

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Tunneling through a system with two discrete electron levels coupled by electron-phonon interaction is considered. The interplay between elastic and inelastic tunneling channels is analyzed not only for weak electron-phonon coupling but also for strong coupling in resonant case. It is shown that intensity and width of peaks in tunneling conductivity is strongly influenced by non equilibrium effects.

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Recently great attention was paid to both experimental and theoretical investigations of kinetic processes in nanostructures, in connection with possible fabrication of electronic devices of ultra small size. Electron-phonon interaction is one of the important effects which influence transport properties of various nanostructures, including structures with quantum dot or a single molecule placed in tunneling contact between the leads. From the other hand, scanning tunneling microscopy (STM) investigation of single molecules on a surface is a powerful method of diagnostics of electron structure changes of adsorbed molecules [1]. One of the important characteristics of these changes is modification of vibrational modes of adsorbed molecule. Thus inelastic tunneling measurements can give information about the type of molecule bounding to the surface. Electron-phonon interaction is also very important in STM experiments, concerning problem of single atomic (molecule) switch, in which atoms or molecules are transferred from one state to another one by a tunneling current [2]. The development of theoretical description should clarify the role of electron-phonon interaction in kinetic processes in nanostructures and help the further progress in this field.

In a number of papers electron-phonon interaction in tunneling structures is described within the most simple model for intermediate system with a single electron level coupled with a phonon mode ([3 – 5]). If interaction with the leads is omitted, then this model has an exact solution [6]. But this model can not be derived consistently from many-body theory of condensed matter (see e.g. [7]) and thus seems to be unrealistic.

We discuss a different model, which can be argued to be more physically justified for various systems, with electron transitions between two levels, accompanied by emission or absorption of a phonon (vibrational quantum). This model allows to describe the interplay between two tunneling channels coupled by the electron-phonon interaction. The influence of such interplay on the tunneling spectra of semiconductor structures with 2D electron layers has been revealed in recent experiments [8]. It was found that tunneling spectra essentially change their shape when intersubband electron transition energy was close to the LO-phonon frequency.

In the present paper we consider the simplest system of this kind, which is described by the Hamiltonian of the following type:

$$\hat{H} = \hat{H}_{\rm dot} + \hat{H}_{\rm tun} + \hat{H}_0. \quad (1)$$

The part $\hat{H}_{dot}$ corresponds to the intermediate subsystem in which we take into account two localized states and induced by electron-phonon interaction transitions between these two states:

$$\hat{H}_{\rm dot} = \sum_{i=1,2} \varepsilon_i a_i^+ a_i + g(a_1^+ a_2 + a_2^+ a_1)(b + b^+) + \omega_0 b^+ b, \quad (2)$$

where ε_i corresponds to discrete levels in quantum dot (or two electron states in molecule), ω_0 – optical phonon frequency (or molecule vibrational mode) and g – is electron-phonon coupling constant. Tunneling transitions from the intermediate system are included in

$$\hat{H}_{\rm tun} = \sum_{\mathbf{p},i=1,2} T_{\mathbf{p},i}(c_{\mathbf{p}}^+ a_i + \text{h.c.}) + \sum_{\mathbf{k},i=1,2} T_{\mathbf{k},i}(c_{\mathbf{k}}^+ a_i + \text{h.c.}). \quad (3)$$

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And free electron spectrum in left and right electrodes ($\mathbf{k}$ and $\mathbf{p}$) includes the applied bias V:

$$\hat{H}_0 = \sum_{\mathbf{k}} (\varepsilon_k - \mu) c^+_{\mathbf{k}} c_{\mathbf{k}} + \sum_{\mathbf{p}} (\varepsilon_p - \mu - eV) c^+_{\mathbf{p}} c_{\mathbf{p}}. \quad (4)$$

Operators $c_{\mathbf{k}}, c_{\mathbf{p}}$ correspond to electrons in the leads and a_i - to electrons at the localized states of intermediate system with energy ε_i.

By means of Keldysh diagram technique the tunneling current through such structure is determined as:

$$I(V) = \sum_{\mathbf{k}, i=1,2} \int d\omega T_{\mathbf{k}i} (G^<_{i\mathbf{k}} - G^<_{\mathbf{k}i}). \quad (5)$$

The expression for the tunneling current can be rewritten in the form, containing only Green functions of the intermediate subsystem [9, 10] :

$$I(V) = 2 \sum_{i=1,2} \gamma^k_i \int [2\mathrm{Im}\, G^R_i(\omega) n^0_k(\omega) - i G^<_{ii}(\omega)] d\omega, \quad (6)$$

where $\gamma^k_i = T_{\mathbf{k}i} T_{i\mathbf{k}} \nu_{\mathbf{k}}(\omega)$, $\nu_{\mathbf{k}}(\omega)$ is the density of states in the left electrode and $n^0_k(\omega)$ - is equilibrium filling number in the left electrode.

The main problem is to calculate the Green functions for the intermediate system in the presence of both the tunneling coupling and electron-phonon interaction. The equations for $G^{R,A}_{i,j}$ are now coupled with the equations for $G^<_{i,j}$ due to electron-phonon interaction. The equations for $G^{R,A}_{i,j}$ as usual describes the modifications of electron spectral functions (density of states), but in this case they depend on electron filling numbers, determined by the kinetic processes. For arbitrary ratio of the electron-phonon coupling constant g to the tunneling rates γ this is a strongly correlated system which can not be solved exactly. The situation reminds widely discussed Hubbard-Anderson model very much.

The theory can be developed in two limiting cases $g \ll \gamma$ and $g \gg \gamma$. In the present work we shall not discuss nonequilibrium phonon effects and suppose that phonon subsystem remains unperturbed by the tunneling processes.

In the limit of weak electron-phonon interaction, $g \ll \gamma$, we can calculate $\mathrm{Im}\, G^R_i(\omega)$ and $G^<_{ii}(\omega)$ as perturbation series in g. The first term looks like:

$$\begin{aligned} G^<_{11}(\omega) = G^R_1(\omega)\Sigma^R_{11}(\omega) G^<_1(\omega) - \\ - G^R_1(\omega)\Sigma^<_{11}(\omega) G^A_1(\omega) + G^<_1(\omega)\Sigma^A_{11}(\omega) G^A_1(\omega), \end{aligned} \quad (7)$$

$$G^R_{11}(\omega) = G^R_1(\omega)\Sigma^R(\omega) G^R_1(\omega). \quad (8)$$

All the Green functions here are calculated for the tunneling problem without electron phonon interaction:

$$\begin{aligned} G^R_i(\omega) &= \frac{1}{\omega - \varepsilon_i + i(\gamma^p_i + \gamma^k_i)}, \\ G^<_i(\omega)) &= -2i n_i(\omega) \mathrm{Im}\, G^R_i(\omega), \end{aligned} \quad (9)$$

where non equilibrium filling number $n_i(\omega) = (\gamma^p_i n^0_p(\omega) + \gamma^k_i n^0_k(\omega)) / (\gamma^p_i + \gamma^k_i)$, and $\Sigma(\omega)$ is the simplest self energy part due to the electron-phonon interaction:

$$\begin{aligned} \Sigma^R_{11}(\omega) = i g^2 \int [D^R(\omega') G^<_2(\omega - \omega') + \\ + D^>(\omega') G^R_2(\omega - \omega')] d\omega', \end{aligned} \quad (10)$$

$$\Sigma^<_{11}(\omega) = -i g^2 \int D^<(\omega') G^<_2(\omega - \omega') d\omega'. \quad (11)$$

The first order contribution to the tunneling current can be divided into two different parts: $I = I_1 + I_2$. The first one corresponds to small changes in electron density of states due to electron-phonon interaction and is described simply by some corrections to G^R_{ii}:

$$\begin{aligned} I_1 = \int \Bigg[\frac{\gamma^k_1 \gamma^p_1}{(\gamma^p_1 + \gamma^k_1)} 2\mathrm{Im}\, G^{(1)R}_{11}(\omega) + \\ + \frac{\gamma^k_2 \gamma^p_2}{(\gamma^p_2 + \gamma^k_2)} 2\mathrm{Im}\, G^{(1)R}_{22}(\omega) \Bigg] \left(n^0_k(\omega) - n^0_p(\omega)\right) d\omega. \end{aligned} \quad (12)$$

The second contribution is more interesting, because it describes inelastic tunneling processes with emission or absorption of a phonon:

$$\begin{aligned} I_2 = 4\pi g^2 \frac{\gamma^k_1 \gamma^p_2 - \gamma^k_2 \gamma^p_1}{(\gamma^p_1 + \gamma^k_1)^2 (\gamma^p_2 + \gamma^k_2)^2} \times \\ \times \int d\omega \mathrm{Im}\, G^R_{11}(\omega) (n^0_k(\omega) - n^0_p(\omega)) \times \\ [\mathrm{Im}\;\, G^R_{22}(\omega + \omega_0)(\gamma^k_1 \gamma^p_2 n^0_p(\omega + \omega_0) - \\ - \gamma^k_2 \gamma^p_1 n^0_k(\omega + \omega_0) + (\gamma^k_1 \gamma^p_2 - \gamma^k_2 \gamma^p_1) N(\omega_0)) - \\ - \mathrm{Im}\, G^R_{22}(\omega - \omega_0)(\gamma^k_1 \gamma^p_2 n^0_p(\omega - \omega_0) - \\ - \gamma^k_2 \gamma^p_1 n^0_k(\omega - \omega_0) + (\gamma^k_1 \gamma^p_2 - \gamma^k_2 \gamma^p_1) N(-\omega_0))]. \end{aligned} \quad (13)$$

The sign of this additional correction to the total current depends on the ratio between four tunneling rates $\gamma^k_1, \gamma^p_1, \gamma^k_2 \gamma^p_2$. The physical reason why electron-phonon interaction can either increase or decrease the tunneling current is connected with interference effects between direct and indirect tunneling channels. It becomes more obvious if we notice that:

$$\frac{\gamma_1^k\gamma_2^p-\gamma_2^k\gamma_1^p}{(\gamma_1^p+\gamma_1^k)(\gamma_2^p+\gamma_2^k)}(n_k^0(\omega)-n_p^0(\omega))=n_1(\omega)-n_2(\omega).$$

So the inelastic processes increase the total current if the inverse population of our two-level system appears due to elastic tunneling. Two examples, which demonstrate the enhancement or suppression effects in the tunneling conductivity are shown in Fig.1.

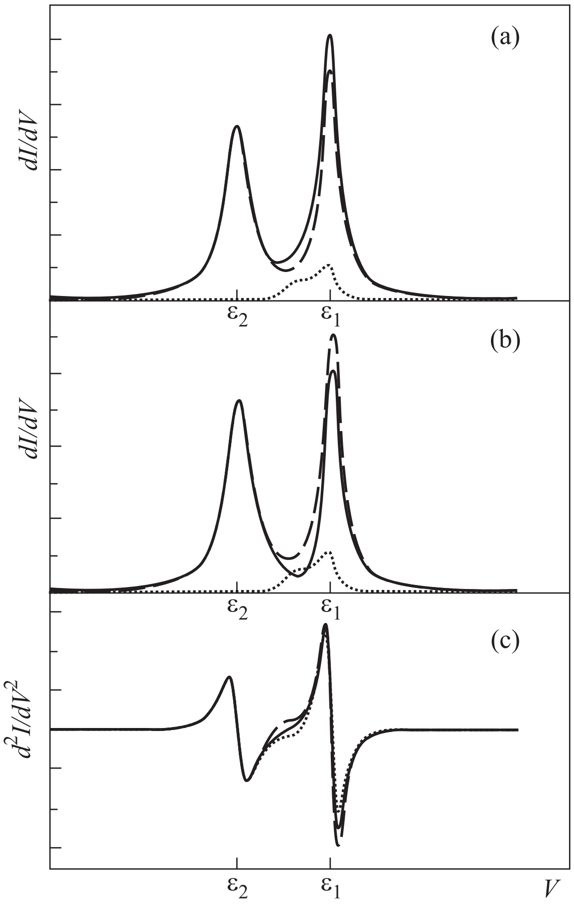

Fig.1. Tunneling conductivity spectra for weak electron phonon coupling. Elastic, inelastic contributions and total tunneling conductivity are shown in Fig.(a), (b) by dashed, dotted and solid curves respectively. Values of the parameters are: $\varepsilon_1-\varepsilon_2=1, \omega_0=0.6, g=0.4$. A) $\gamma_1^k\gamma_2^p \gg \gamma_2^k\gamma_1^p$; B) $\gamma_1^k\gamma_2^p \ll \gamma_2^k\gamma_1^p$ Phonon induced structure is more pronounced in d^2I/dV^2 as it is shown in Fig.(c) by dashed line for the case "a" and dotted line – for the case "b", solid line corresponds to elastic part of the current

In the limit of small tunneling coupling to the leads so, that all $\gamma \ll g$, we use a perturbation treatment of tunneling processes in small parameter γ/g. For the most interesting case, when phonon frequency is close to the resonance with electron transition energy $\varepsilon_1-\varepsilon_2 \simeq \omega_0$ we can retain in the Hamiltonian only the following interactions (similar to the rotating wave approximation for two-level systems) :

$$\hat{H}_{el-phon}=g(a_1^+a_2b+a_2^+a_1b^+)$$

as the most important.

Direct calculation of dot (molecule) Green functions G_{11}^R, G_{22}^R based on Heisenberg equations for isolated dot (molecule) leads to a system of equations:

$$(\omega-\varepsilon_1)G_{01}^R-gG_{2b1}^R=1,$$
$$(\omega-\varepsilon_2-\omega_0)G_{2b1}^R-gG_{2n1}^R=0,$$
$$(\omega-\varepsilon_1)G_{2n1}^R-g(N+1)G_{2b1}^R=1-n_2+N.$$

Where new functions G_{2b1}^R, G_{2n1}^R are introduced:

$$G_{2b1}^R=i\theta(t-t')\langle[a_2(t)b(t),a_1^+(t')]\rangle,$$

$$G_{2n1}^R=i\theta(t-t')\langle[a_1(t)(b(t)b^+(t)-a_2^+(t)a_2(t)),a_1^+(t')]\rangle.$$

N and n_2 are the number of phonons and electrons at the second level respectively. At the third step the mean value of phonon filling number is decoupled from the average $< a_2b(1+b^+b), a_1^+ > \longrightarrow < a_2b, a_1^+ > (N+1)$. After this approximation was done the retarded Green function is determined:

$$G_{01}^R=\frac{1}{(\omega-\varepsilon_1)}\times$$
$$\times\left[1+\frac{g^2(N+1-n_2)}{((\omega-\varepsilon_1)(\omega-\varepsilon_2-\omega_0)-g^2(N+1))}\right]. \quad (14)$$

The same procedure for the second level gives:

$$G_{02}^R=\frac{1}{(\omega-\varepsilon_2)}\times$$
$$\times\left[1+\frac{g^2(N+n_1)}{((\omega-\varepsilon_2)(\omega-\varepsilon_1+\omega_0)-g^2(N+1))}\right]$$

We see, that new poles in the Green functions appeared, which corresponds to splitting of resonantly close two pairs of levels: ε_1 and $\varepsilon_2+\omega_0$ as well as ε_2 and $\varepsilon_1-\omega_0$. We can expect the appearance of three resonant peaks in the local density of states near ε_1 and ε_2. Note that the energies of two split states depend on the temperature through the phonon filling number N.

Now we treat as the first order perturbation the self energy parts of two kinds, appeared due to the tunneling coupling with the leads (see Fig.2.)

Tunneling current is determined by the self energies as

$$I=2\sum_{i=1,2}\gamma_i^k\int[G_i^R(\omega)(-i\Sigma_{ii}^<(\omega))G_i^A(\omega)+$$
$$+n_k^0(\omega)G_i^R(\omega)\mathrm{Im}\Sigma_{ii}^R(\omega)G_i^A(\omega)]d\omega$$

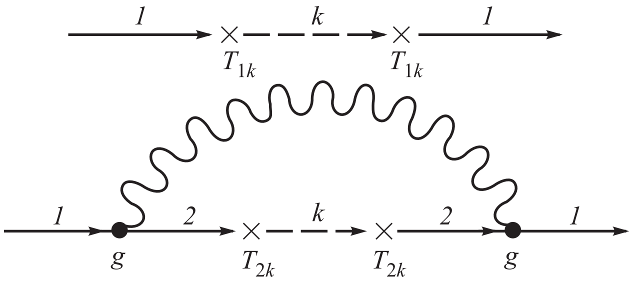

Fig.2. Two first order diagrams for the self energy part Σ_{11}. Dashed line denotes electron Green function in the leads, wavy line – phonon Green function

The contribution of the first diagram is rather simple

$$I_1 = \int \left[\gamma_1^k \gamma_1^p G_1^R(\omega) G_1^A(\omega) + \gamma_2^k \gamma_2^p G_2^R(\omega) G_2^A(\omega)\right] \times$$
$$\times \left(n_k^0(\omega) - n_p^0(\omega)\right) d\omega.$$

The contribution described by the second diagram reduces in the first non vanishing order in $(\gamma/g)^2$ to the following form

$$I_2 = g^2 \int d\omega |G_1^R(\omega)|^2 |G_2^R(\omega - \omega_0)|^2 \left(n_k^0(\omega) - n_p^0(\omega)\right) \times$$
$$\times(\gamma_1^k \gamma_2^p (n_p^0(\omega - \omega_0) - N(\omega_0) - 1)+$$
$$+\gamma_2^k \gamma_1^p (n_k^0(\omega - \omega_0) - N(\omega_0) - 1)).$$

It is important that functions G_i^R are determined with the help of Eq. (14):

$$[G_i^R]^{-1} = [G_{0i}^R]^{-1} + i(\gamma_i^k + \gamma_i^p).$$

This expression allows us to estimate the effective width of each of the three resonant peaks near the initial electron levels. It is remarkable that broadening of these peaks is different and depends on nonequilibrium electron numbers, thus it can be tuned by changing the parameters of the tunneling contact. The peak, corresponding to ε_1 has the effective width: $\Gamma_1^0 = (\gamma_1^k + \gamma_1^p) n_2/(N+1)$. And two split peaks near it have the width $\Gamma_1^{\pm} = (\gamma_1^k + \gamma_1^p)(N + 1 - n_2)/2(N+1)$. The same values for the second set of peaks near ε_2 are: $\Gamma_2^0 = (\gamma_2^k + \gamma_2^p)(1 - n_1)/(N+1), \Gamma_2^{\pm} = (\gamma_2^k + \gamma_2^p)(N + n_1)/2(N+1)$.

Some tunneling conductivity curves for zero temperature ($N = 0$) are shown in Fig.3. In this figure we consider the case when both levels ε_1 and ε_2 lie above the Fermi level at zero applied bias voltage.

Let us point out the most interesting features of the tunneling conductivity behavior in this case. For the elastic channel in the tunneling conductivity spectra nonequilibrium narrowing of two split peaks near ε_2 is always clearly seen, while the peak at initial energy ε_2 is broadened in a usual way by the tunneling rates. In bias range close to ε_1 we can distinguish two different cases, dependent on the ratio between the tunneling rates, because it determines the filling number of more deep state ε_2. For $\gamma_2^k \ll \gamma_2^p$ two split peaks near ε_1 are narrow and

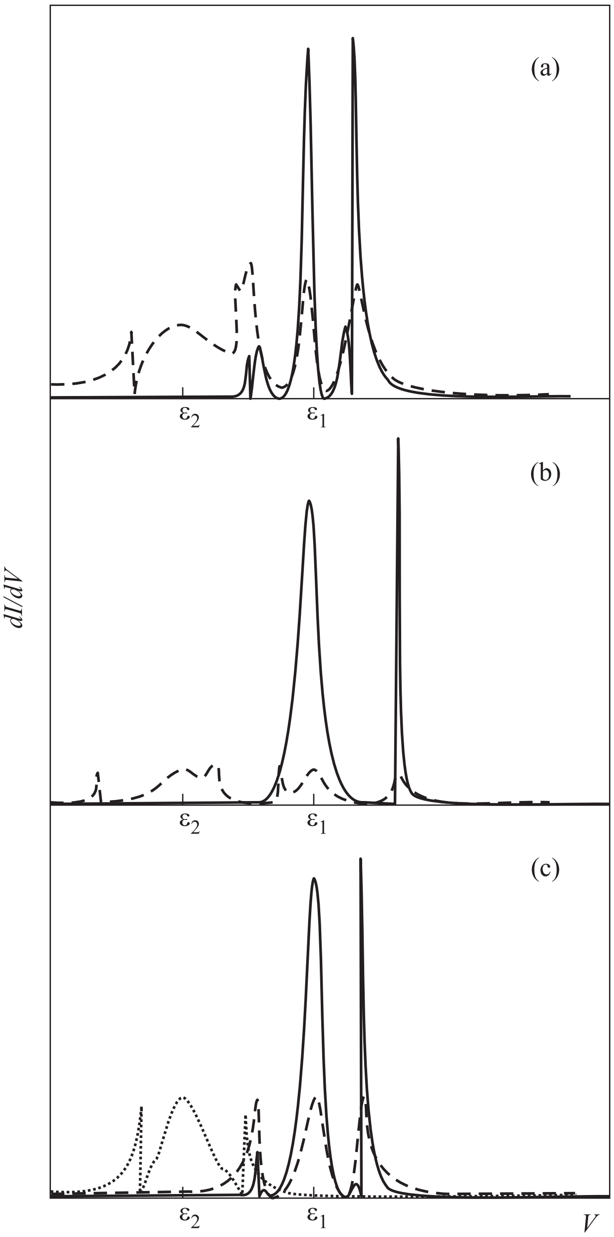

Fig.3. Tunneling conductivity spectra for strong resonant electron phonon coupling. Elastic and inelastic contributions to the tunneling conductivity are shown by dashed and solid curves respectively. Chosen values of the parameters are: $\varepsilon_1 - \varepsilon_2 = 1, \omega_0 = 0.9, g = 0.4$. (a) $\gamma_2^k = 0.3, \gamma_2^p = 0.06$; (b) $\gamma_2^k = 0.06, \gamma_2^p = 0.3$; (c) $\gamma_2^k = \gamma_2^p = 0.1$. In figure (c) elastic tunneling current through each electron level is shown separately by dotted and dashed curves

tunneling conductivity peak at bias voltage equal to initial level ε_1 has usual tunneling width (Fig.3.).

For $\gamma_2^k \gg \gamma_2^p$ vice versa the split peaks are broadened up to the usual tunneling width and peak at bias voltage equal to initial level ε_1 becomes narrow. For strong enough electron-phonon coupling contribution of inelastic channel to the tunneling conductivity can strongly exceed the elastic channel contribution at certain bias close to ε_1. The tunneling conductivity peaks of inelastic channel are narrow due to non equilibrium effects.

At high temperature, when $N \gg 1$, the shape of the tunneling conductivity curves are not so sensitive to the ratio between different tunneling rates. One should observe very narrow peaks at ε_1 and ε_2, while the broadening of split peaks would be always equal to one half of the usual tunneling width.

Conclusions. In the weak electron phonon coupling limit we revealed that interference effects between various channels can lead either to some increase of the total current or to its suppression. The sign of the resulting effect depends on the ratios between the tunneling rates.

For too large tunneling rates observation of inelastic peaks in the tunneling conductivity spectra is hardly possible.

For strong electron-phonon coupling three peaks can arise in the tunneling conductivity spectra near each electron level in resonant situation. Non equilibrium narrowing of these peaks at certain ratio between the tunneling rates is found out. The positions of satellite peaks in this case are determined not only by the phonon frequency ω_0, but also by the electron phonon coupling constant and the temperature.

The important feature of the system under consideration is that we can change the relative intensity and width of elastic and inelastic peaks by tuning the tunneling coupling of intermediate system with the leads.

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