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Tunneling current induced phonon generation in nanostructures

P. I. Arseyev[1)], *N. S. Maslova*[†]

P.N.Lebedev Physical Institute of RAS, 119991 Moscow, Russia

[†]*Department of Physics, Moscow State University, 119992 Moscow, Russia*

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We analyze generation of phonons in tunneling structures with two electron states coupled by electron-phonon interaction. The conditions of strong vibration excitations are determined and dependence of non-equilibrium phonon occupation numbers on the applied bias is found. For high vibration excitation levels self consistent theory for the tunneling transport is presented.

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Tunneling current induces generation of phonons (or vibrational quanta for a molecule) leading to effective "heating" of the phonon subsystem in any real system with electron-phonon interaction. In scanning tunneling microscopy experiments this effect may induce motion, dissociation or desorption of adsorbed molecules thus allows single molecule manipulation on a surface [1 – 3]. The main purpose of the present work is to reveal the conditions for strong phonon generation as well as for its suppression. We also investigate the influence of strong phonon generation on tunneling conductivity behavior.

In our previous work [4] we analyzed modifications of tunneling current using a model in which electron-phonon interaction leads to transitions between two electron levels. In the present paper we demonstrate that in this system tunneling current can induce strong phonon generation. The mechanism of this enhanced phonon heating is closely connected with the presence of at least two electron states in a molecule or a quantum dot(QD), and in a single level model, widely discussed in literature ([5 – 7]), this mechanism is absent.

We consider a tunneling system, which is described by the Hamiltonian of the following type:

$$\widehat{H} = \widehat{H}_{\rm dot} + \widehat{H}_{\rm tun} + \widehat{H}_0. \tag{1}$$

The part $\widehat{H}_{\rm dot}$ corresponds to a QD or a molecule with two localized electron states taking into account. Electron-phonon interaction leads to transitions between these two states.

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

where ε_i corresponds to discrete levels in quantum dot (or two electron states in molecule) and we adopt that $\varepsilon_1 > \varepsilon_2$, 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 $\widehat{H}_{\rm tun}$

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

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

$$\widehat{H}_0 = \sum_{\mathbf{k}} (\varepsilon_k - \mu) c_{\mathbf{k}}^+ c_{\mathbf{k}} + \sum_{\mathbf{p}} (\varepsilon_p - \mu - eV) c_{\mathbf{p}}^+ c_{\mathbf{p}}. \tag{4}$$

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 non-equilibrium phonon numbers can be found from Dyson equations for the phonon Green function. To determine phonon occupation numbers we need to solve the Dyson equation for $D^<$ together with the equation for $D^{R(A)}$ which gives:

$$D^< = \frac{\Pi^<}{\Pi^A - \Pi^R}(D^R - D^A). \tag{5}$$

Polarization operators Π are easily calculated in the lowest order in electron-phonon interaction:

$$\Pi^A(\Omega) - \Pi^R(\Omega) = -4ig^2 \times$$
$$\times \int \frac{d\omega}{2\pi} \left[\text{Im} G_2^A(\omega) \text{Im}\, G_1^A(\omega - \Omega)(n_2(\omega) - n_1(\omega - \Omega)) + \right.$$
$$\left. + \text{Im}\, G_1^A(\omega) \text{Im} G_2^A(\omega - \Omega)(n_1(\omega) - n_2(\omega - \Omega)) \right],$$

$$\Pi^<(\Omega) = -4ig^2 \times$$
$$\times \int \frac{d\omega}{2\pi} \left[n_1(\omega)(n_2(\omega - \Omega) - 1) \text{Im}\, G_1^A(\omega) \text{Im}\, G_2^A(\omega - \Omega) + \right.$$

[1)] e-mail: ars@lpi.ru

$$+ n_2(\omega)(n_1(\omega-\Omega)-1)\mathrm{Im}\,G_2^A(\omega)\mathrm{Im}\,G_1^A(\omega-\Omega)\big]\,. \quad (6)$$

All electron Green functions in the above expressions are calculated with full account for tunneling transitions. Thus electron non equilibrium filling numbers n_1, n_2 are determined by the tunneling processes (neglecting electron-phonon interaction) as: $n_i(\omega) = (\gamma_i^p n_p^0(\omega) + \gamma_i^k n_k^0(\omega))/\gamma_i$. Tunneling rates γ are determined as usually by the corresponding tunneling matrix elements $T_{k,(p),i}$ and densities of states $\nu_{k,p}$ of the leads: $\gamma_i^k = T_{k,i}^2\nu_k$, $\gamma_i^p = T_{p,i}^2\nu_p$. Total width of electron levels due to the tunneling coupling is denoted by $\gamma_i = \gamma_i^p + \gamma_i^k$.

Equation (5) together with the relation $D^<(\Omega) = N(\Omega)(D^R(\Omega) - D^A(\Omega))$ leads to the following nonequilibrium phonon numbers:

$$N(\Omega) = \frac{\Pi^<}{\Pi^A - \Pi^R}. \quad (7)$$

Polarization operator $\Pi^<$ can be divided into two parts:

$$\Pi^<(\Omega) = 2i\mathrm{Im}\,\Pi^A N_0(\Omega) + iP^<(\Omega). \quad (8)$$

Which allows separate explicitly the equilibrium Bose-function $N_0(\Omega)$ from nonequilibrium occupation numbers:

$$N(\Omega) = N_0(\Omega) + \Delta N(\Omega), \quad (9)$$

where

$$\Delta N(\Omega) = P^<(\Omega)/2\mathrm{Im}\,\Pi^A. \quad (10)$$

After substitution in $\mathrm{Im}\,\Pi^A$ (Eq.(6)) for n_1, n_2 their explicit expressions in terms of Fermi functions in the leads n_p and n_k a term proportional to the combination $(\gamma_1^k\gamma_2^p - \gamma_2^k\gamma_1^p)$ of the tunneling rates appears in $\mathrm{Im}\,\Pi^A$. The sign of this combination determines the relative population of the two electron levels due to the tunneling current through the system, because the following relation holds:

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

Considering phonon generation processes cases of "normal" $(n_1(\omega) - n_2(\omega) < 0)$ and inverse $(n_1(\omega) - n_2(\omega) > 0)$ population for the two levels with $\varepsilon_1 > \varepsilon_2$ drastically differ from each other. In the case of normal occupation phonon generation is rather weak. Typical dependence of phonon occupation numbers on bias voltage calculated by the help of Eq. (10) is depicted in Fig.1. Maximum value of nonequilibrium phonon numbers does not exceed several units. Increasing of temperature smears fine structure of this dependence and always decreases maximum values of phonon generation.

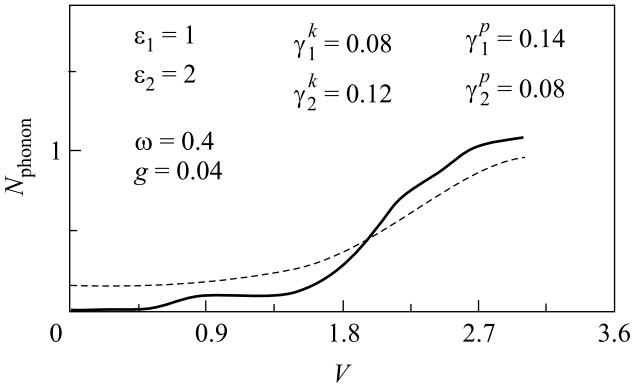

Fig.1. N_{phonon} versus applied bias for weak generation regime. Solid line corresponds to zero temperature, and dashed line – to $T = 0.2$

On the contrary in the case of inverse occupation at some bias voltage the nonequilibrium generation infinitly increases. The divergence of nonequilibrium phonon occupation numbers occurs because at some bias voltage $\mathrm{Im}\,\Pi^A$ given by Eq.(6) passes through zero, changing its sign. In order to describe correctly the threshold of phonon generation it is necessary to take into account higher order terms which allows to take into account nonlinear in phonon occupation numbers effects. For "normal occupation" case $\mathrm{Im}\,\Pi^A$ is always positive for any voltage and the Eq. (10) is sufficient to calculate the non-equilibrium phonon occupation numbers.

In a tunneling system with inverse electron level occupation one should self-consistently take into account modifications of electron Green functions due to electron-phonon interaction together with nonequilibrium phonon numbers. The first corrections to the electron Green functions and to the electron-phonon vertex of the order of $g^2D(\Omega)$ (where D is dressed phonon Green function) is shown in Fig.2.

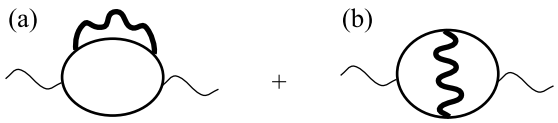

Fig.2. Diagrams for $\Pi^{(1)}$

Diagrams in Fig.2a describe the part of $\mathrm{Im}\,\Pi^{(1)A}$, which is connected with the corrections to electron Green functions. They can be analytically expressed by Eq. (6), with g^2 order corrections either to $\mathrm{Im}\,G_1^A$, or to $\mathrm{Im}\,G_2^A$, or to occupation numbers n_1 and n_2. In the present problem vertex corrections to polarization op-

erator shown in Fig.2b are not so small as it is usually supposed for bulk electron phonon interaction, and also should be taken into account. As we shall discuss below, in the weak coupling regime the width of the phonon lines remains narrow enough even in the presence of strong phonon generation. This allows us to use δ-function approximation for the phonon spectral function $\mathrm{Im}\, D^R$ throughout the calculations.

It is important that some corrections appeared in $\mathrm{Im}\,\Pi^{(1)A}$ are proportional to the nonequilibrium phonon numbers $N(\omega_0)$. These terms ensure nonlinear limiting of phonon generation. In the following we retain only these terms, because the rest part of $\mathrm{Im}\,\Pi^{(1)A}$, independent of $N(\omega_0)$, gives only very small shift of threshold voltage for strong phonon generation and can be omitted.

Integrating Eq. (7) over Ω near ω_0 we get the equation for self-consistent calculation of $N_{ph}(\omega_0) \simeq \simeq \int d\Omega N(\Omega)\mathrm{Im}\, D^A(\Omega)$:

$$N(\omega_0) = \frac{\Pi^<(\omega_0)}{2i(\mathrm{Im}\,\Pi^A(\omega_0) + \mathrm{Im}\,\Pi^{(1)A}(\omega_0))}. \qquad (11)$$

There is no need to calculate corrections to the function $\Pi^<(\omega_0)$, because it is always positive and never becomes close to zero. So all the highest order corrections are inessential in this case.

Finally we can rewrite Eq. (11) in the following form:

$$N(\omega_0) = \frac{N_0(\omega_0)2\mathrm{Im}\,\Pi^A(\omega_0) + P^<(\omega_0)}{2\mathrm{Im}\,\Pi^A(\omega_0) + g^2N(\omega_0)A(\omega_0)}. \qquad (12)$$

Where $P^<$ and $\mathrm{Im}\,\Pi^A$ are determined by the equations (8) and (6). All the essential second order corrections to $\mathrm{Im}\,\Pi^A$ proportional to $N(\omega_0)$ are now rewritten as $g^2N(\omega_0)A(\omega_0)$. The function $A(\omega_0)$ depends on applied bias voltage through electron filling numbers in the leads. Complete expression for $A(\omega_0)$, obtained from diagrams in Fig.2. is too complicated, and we do not reproduce it here.

For the normal electron level population ($g^2A(\omega_0) \ll \mathrm{Im}\Pi^A(\omega_0)$ in all bias range) Eq. (12) gives only small corrections to the first order Eq. (10) for nonequilibrium phonon numbers.

For inverse population the appearance of correction $g^2A(\omega_0)$ is crucial, because $\mathrm{Im}\,\Pi^A(\omega_0)$ changes its sign and is equal to zero at some threshold value of applied voltage. It was found out that function $A(\omega_0)$ also changes its sign (sometimes not once) being positive at large bias. Moreover the bias value at which $A(\omega_0)$ goes from negative to positive values last time is very close to the threshold value for $\mathrm{Im}\,\Pi^A(\omega_0)$. This is the reason why we should retain all the corrections of the second order to polarization operators in order to get the reasonable accuracy of calculations. It was checked by direct numerical investigations, that for some parameters of the contact only vertex corrections ensure the validity of Eq. (12) in all bias range.

For large bias voltages, greater than the threshold value, $\mathrm{Im}\,\Pi^A$ becomes large negative value. So we get from Eq. (12) the following expression for the phonon occupation numbers:

$$N(\omega_0) \simeq \frac{2|\mathrm{Im}\,\Pi^A(\omega_0)|}{g^2A(\omega_0)}. \qquad (13)$$

For voltages beyond the threshold and if $|\varepsilon_1 - \varepsilon_2 - \omega_0| \geq \geq \gamma_1 + \gamma_2$ the value of $\mathrm{Im}\,\Pi^A(\omega_0)$ can be estimated as:

$$|\mathrm{Im}\Pi^A(\omega_0)| \simeq \frac{g^2}{\gamma_1\gamma_2}(\gamma_1^k\gamma_2^p - \gamma_2^k\gamma_1^p)\frac{\gamma_1+\gamma_2}{(\varepsilon_1-\varepsilon_2-\omega_0)^2}. \qquad (14)$$

For the resonant case denominator $(\varepsilon_1 - \varepsilon_2 - \omega_0)$ is replaced by $(\gamma_1 + \gamma_2)$ in this expression. The value of A for large voltages (saturation regime) can be estimated as:

$$A(\omega_0) \simeq \frac{g^2}{\gamma_1\gamma_2}\frac{\gamma_1+\gamma_2}{(\varepsilon_1-\varepsilon_2-\omega_0)^2}. \qquad (15)$$

Therefore maximum occupation numbers in saturation regime at high applied voltages are:

$$N_{\max} \simeq \frac{(\gamma_1^k\gamma_2^p - \gamma_2^k\gamma_1^p)}{g^2}. \qquad (16)$$

Since we consider the case of weak electron-phonon interaction, $g \ll \gamma_1, \gamma_2$, phonon occupation numbers are large, which means strong phonon generation.

Now let us return to the problem of self-consistency of the presented second order calculations. The broadening of electron levels due to electron-phonon interaction is determined by $\mathrm{Im}\,\Sigma^A_{1,2}$ represented by the first diagram in Fig.3 (see also [4]). Our approximation remains

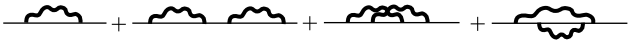

Fig.3. Corrections to electron Green function

valid until $\mathrm{Im}\,\Sigma^A_{1,2}$ is less, than the broadening of electron levels $\gamma_{1,2}$ due to the tunneling coupling. For large phonon occupation numbers the main term of $\mathrm{Im}\,\Sigma^A_1$ is:

$$\mathrm{Im}\,\Sigma^A_1(\omega) = g^2N(\omega_0)(\mathrm{Im}\,G^A_2(\omega-\omega_0) + \dots). \qquad (17)$$

Thus $N(\omega_0)$ is limited by inequality:

$$\max \mathrm{Im}\,\Sigma^A_1 \simeq \frac{g^2}{\gamma_2}N(\omega_0) < \gamma_1. \qquad (18)$$

From Eq.(18) the limit of validity of our approximation is:

$$\max N < \gamma_1\gamma_2/g^2. \tag{19}$$

This value of N_{max} from Eq. (16) is just of the same order. So at large values of applied bias we, strictly speaking, work at the limit of applicability of suggested scheme but never go beyond this limit.

The phonon line width is determined by the imaginary part of the polarization operator Π^A. Omitting second order corrections, maximum value of $\mathrm{Im}\,\Pi^A \simeq \simeq g^2/(\gamma_1+\gamma_2)$ (see (14)) is much smaller than the phonon frequency ω_0. Second order corrections to $\mathrm{Im}\,\Pi^A$ do not change substantially this value, even for great nonequilibrium phonon numbers. In accordance with Eqs. (18), (16) maximum possible changes of electron Green functions are of the order of themselves. Thus corrections to polarization operators can result only in some numerical coefficient of the order of unity. This means that even in the saturation regime with large nonequilibrium phonon numbers the width of the phonon line remains small.

Tunneling current through two-level system can be expressed as [4, 8]:

$$I(V) = 2\sum_{i,j=1,2}\gamma_i^k\int[\mathrm{Im}\,G_i^A(\omega)n_k^0(\omega) - iG_i^<(\omega)]d\omega. \tag{20}$$

Corrections to the current are thus determined by the corrections to the electron Green functions $G^A_{1,2}$ and $G^<_{1,2}$. First order corrections corresponding to the first diagram in Fig.3 with equilibrium phonon Green function were calculated in the previous paper [4]. If we take into account nonequilibrium changes of the phonon occupation numbers, we should consider four diagrams, depicted in Fig.3.

It is worth to remark that just these types of the electron self-energy corrections are consistent with phonon polarization operators, depicted in Fig.2. The both sets of diagrams originate from the the same generating functional.

This self-consistency means, that corresponding Ward identities are satisfied, and charge conservation in the tunneling processes is automatically fulfilled. Note that all electron Green functions in all polarization operators and self-energy parts are calculated omitting the electron-phonon interaction. An attempt to use self-consistent Born approximation for the electron GF leads to violation of charge conservation in this problem and artificial symmetrization is needed to restore current continuity ([9, 10]).

But we can neglect the contribution from the three last diagrams in Fig.3. in the following cases:

a) If there is no inverse electron population due to the tunneling current, then phonon numbers are small, and second order diagrams have small parameter $g^2/\gamma_1\gamma_2$.

b) If inverse population appears, but phonon frequency is far from the resonance between electron levels: $|\varepsilon_1-\varepsilon_2-\omega_0| \geq \gamma_1, \gamma_2$. Then second order diagrams have small parameter $\gamma_i/|\varepsilon_1-\varepsilon_2-\omega_0|$. In this case phonon numbers are large $N \gg 1$, so the first correction with full phonon function strongly differs from equilibrium result.

c) If inverse population appears and phonon frequency is almost in resonance with electron transition energy $|\varepsilon_1-\varepsilon_2-\omega_0| \leq \gamma_1, \gamma_2$, then we can restrict ourself to the first term only near the threshold voltage, until phonon numbers are not too large. For high voltages in saturation regime in this case we can not use perturbation theory, because corrections to electron Green functions become of the order of zero order Green function. We should sum up not only second order diagrams depicted in Fig.3, but all the higher order terms as well.

So we calculate the corrections to the tunneling current using only the first term in Fig.3 (one-loop diagram) but with full nonequilibrium phonon green function, taking into account that in resonant case c) it makes sense not far beyond the threshold of strong generation.

In addition to the tunneling current calculated in [4] there exists a correction to Eq. (20) proportional to nonequilibrium phonon numbers:

$$\begin{aligned}\Delta I = 2g^2N(\omega_0)\int\frac{d\omega}{2\pi}\Bigg[\Bigg[\frac{\gamma_1^k\gamma_1^p}{\gamma_1}\mathrm{Im}\{(G_1^{(1)A}(\omega))^2\}\times\\ \times(\mathrm{Re}\,G_2^A(\omega-\omega_0)+\mathrm{Re}\,G_2^A(\omega+\omega_0))+\\ +\mathrm{Re}\{(G_1^{(1)A}(\omega))^2\}(\mathrm{Im}\,G_2^A(\omega-\omega_0)+\\ +\mathrm{Im}\,G_2^A(\omega+\omega_0))\Bigg](n_k^0(\omega)-n_p^0(\omega))-\\ -\frac{\gamma_1^k\gamma_2^p-\gamma_2^k\gamma_1^p}{\gamma_1\gamma_2}\times(n_1(\omega)-n_2(\omega-\omega_0))\times\\ \times\mathrm{Im}\,G_1^A(\omega)\mathrm{Im}\,G_2^A(\omega-\omega_0)+(1\longleftrightarrow 2)\Bigg].\end{aligned} \tag{21}$$

Since $N(\omega_0)$ depends on applied bias increasing rapidly at some threshold voltage, new peculiarities connected with dN/dV can appear in tunneling conductivity (dI/dV).

This peculiarity is more pronounced if $\omega_0 \geq (\varepsilon_1-\varepsilon_2)$. The peak in dI/dV at phonon generation threshold voltage (when $dN/dV \gg 1$) is clearly seen in Fig.4.

For $\omega_0 \leq (\varepsilon_1-\varepsilon_2)$ nonequilibrium phonon numbers can depend on applied bias non-monotonically (Fig.5). But in this case, in spite of great phonon excitation,

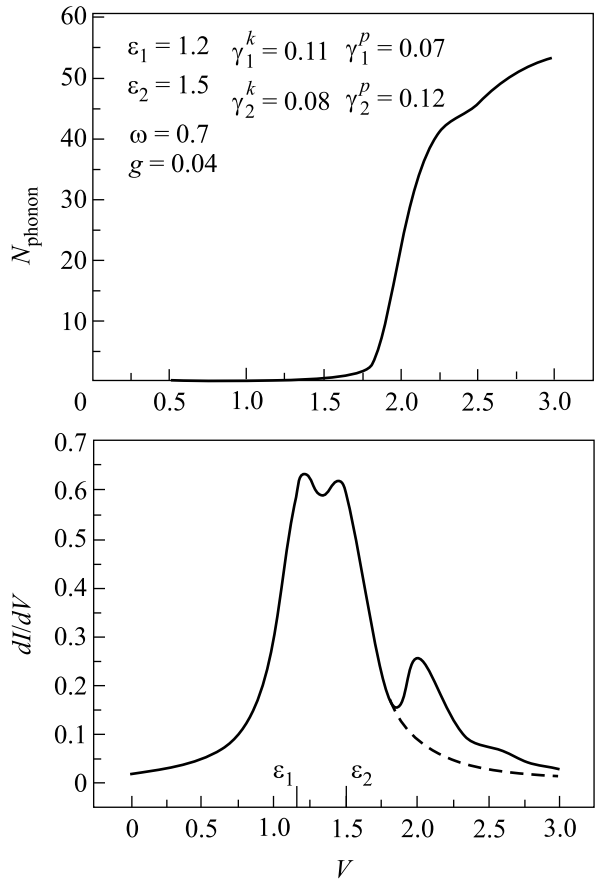

Fig.4. N_{phonon} and dI/dV versus applied bias for strong generation regime. Conductivity without electron-phonon interaction is shown by the dashed line

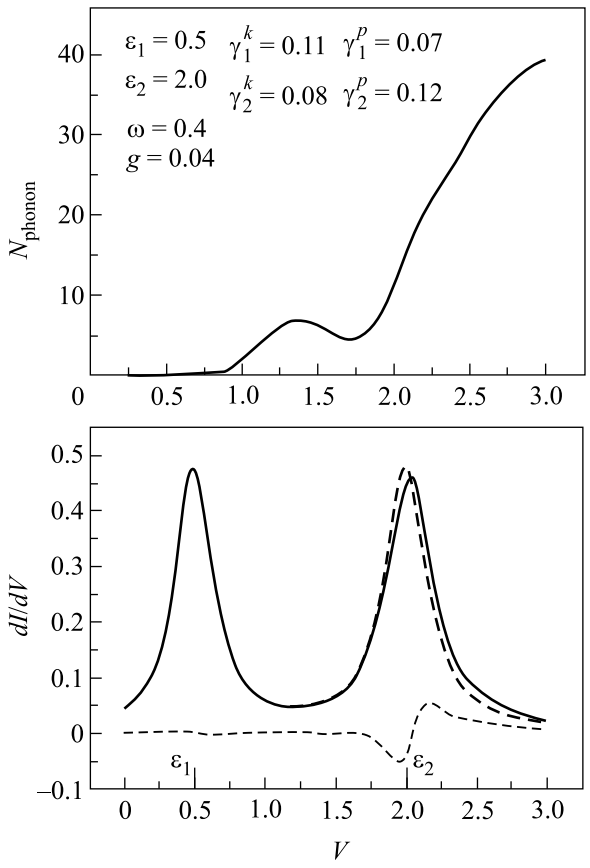

Fig.5. Strong generation regime with non-monotonous behavior of N_{phonon}. Correction to dI/dV (dotted line) is pronounced but is not seen in the total conductivity (solid line)

no definite peculiarities connected with electron-phonon interaction can be observed in tunneling conductivity curves (see Fig.5). Approach to saturation regime for N at large bias is clearly seen in Fig.4, 5. The value of $N_{\max}$ is in agreement with estimation given by Eqs. (16), (19). The obtained values for $N_{\max} \simeq 30-40$ for reasonable junction parameters can hardly be observed in real small systems, because such great overheating should lead to dissociation, desorption of molecules or destruction of quantum dots. In order to deal with real objects at such great intensity of vibration excitation one should take into account phonon anharmonicity and other relaxation processes for the phonons.

In conclusion we point out that intensity of phonon generation can be tuned by changing the parameters of the tunneling junction (which influence the tunneling rates). For inverse population of two-level electron system strong phonon generation takes place which can lead to drastic changes of the properties of nanostructures. Let us note that the problem of suppression of tunneling current induced phonon generation is very important for fabrication of semiconductor cascade lasers based on sequence of tunneling junctions [11]. Generation of optical radiation requires the inverse population of two electron states. But, as we have shown in the present work, in this case strong phonon generation inevitably appears and always competes with optical radiation generation. Using the results of the present paper one could analyze whether it is possible to achieve the threshold of optical generation before strong phonon generation begins changing the parameters of the tunneling system.

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