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Tunneling current induced squeezing of the single-molecule vibrational mode

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A tunneling system with a molecule between the leads was analyzed in the framework of the adiabatic approach. The adiabatic approach allowed us to consider effects of equilibrium ion positions changing and vibrational mode frequency modification due to the electrons tunneling through the molecule localized states. It was demonstrated that vibrational states become squeezed if the tunneling current flows through the molecule. The mean-square displacement of the molecule and the degree of squeezing depend on the system parameters and could be tuned by applied bias voltage. Obtained results demonstrate the possibility of molecule ions' effective "freezing" by the tunneling current.

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I. INTRODUCTION

During the last decade electronic devices have demonstrated significant size reduction which has led to the increasing role of quantum effects. It is quite clear that in nonlinear small size electronic systems there exists a set of quantum states—squeezed states—which have less uncertainty in one quadrature than a coherent state [1]. Realization of such states is a key problem in the development of quantum communication and one of the crucial points in the implementation of quantum information and stability enhancement of quantum electronic devices [2]. Squeezed states are important tools for continuous variable quantum information protocols, as they could be used for generation of the nonclassical states demonstrating reduced noise in one of their quadratures and amplification in another one [3,4]. Their enhanced sensitivity could be also applied in metrology [5–7], e.g., atomic clocks [8], interferometric measurements [9,10], atomic absorption spectroscopy [11], imaging [12,13], and particle tracking in biological systems [14]. Moreover, the appearance of squeezed states strongly changes the dynamics of quantum systems [15–17]. Unfortunately, most of the experiments demonstrating the production and application of squeezed states have been performed in optical [18,19] or mechanical [20–22] systems. The recent advent of quantum information processing by means of superconducting circuits leads to the production of squeezed states at microwave frequencies [23]. Microwave squeezed states could improve measurement sensitivity for qubit state readout [24,25]. In [26] the appearance of squeezed phonon states was described in the superconductor tunneling junction. The appearance of squeezed phonon states was caused by the interaction between phonon subsystems and coherent superconducting electron states. Recently, hybrid systems were realized where a quantum conductor is coupled to a microwave resonator. In solids individual molecules, impurities, or quantum dots are promising candidates to realize squeezed states. A number of experiments were performed with metallic tunneling junctions coupled to quantum dots, nanowires, or two-dimensional electron gas introduced in coplanar cavities [27–30]. The nonequilibrium electron transport through a confined structure could lead to the emission of nonclassical light by the quantum conductor [31,32]. Squeezed light could be also emitted by the tunneling junction in the absence of a cavity [33,34]. Later the possibility for ground-state squeezing and suppression of the spin current short noise were predicted in a hybrid system nonmagnetic conductor–ferromagnet driven by a coherent microwave pump [35,36].

In this paper we analyze the influence of tunneling processes on the molecule vibrational mode states in the normal tunneling junction. The physics presented herein relies upon the adiabatic approximation. We study the appearance of vibrational mode squeezed states caused by the tunneling current and demonstrate that ion mean-square displacement can decrease beyond the quantum limit. Moreover, it is shown that the degree of squeezing could be tuned by the applied bias voltage.

II. THEORETICAL MODEL AND MAIN RESULTS

Let us consider for simplicity a diatomic molecule localized in the tunneling contact (an absorbed molecule on the semiconductor/metal surface. Scheme of the system under investigation is shown in Fig. 1). The complete system Hamiltonian can be written in the following form:

$$\hat{H} = \hat{H}_0 + \hat{H}_{\mathrm{leads}} + \hat{H}_{\mathrm{tun}}, \tag{1}$$

where isolated molecule part $\hat{H}_0$ and tunneling part $\hat{H}_{\mathrm{tun}}$ include electron-phonon interaction and term $\hat{H}_{\mathrm{leads}}$ describes

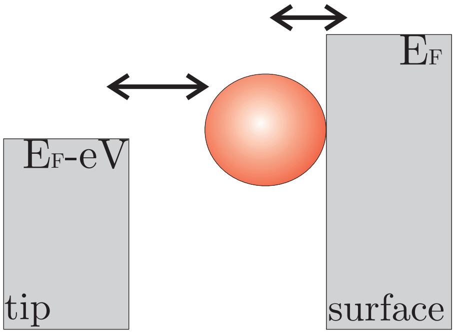

FIG. 1. Scheme of the molecule localized in the tunneling junction formed, for example, by the semiconductor/metal surface and tip of the scanning tunneling microscope.

conduction electrons in the tunneling contact leads:

$$\hat{H}_{\text{leads}} = \sum_k \varepsilon_k \hat{c}_k^+ \hat{c}_k + \sum_p (\varepsilon_p - \text{eV}) \hat{c}_p^+ \hat{c}_p. \tag{2}$$

Here index $k(p)$ labels continuous spectrum states in the tunneling contact leads with the energies $\varepsilon_{k(p)}$. Operators $\hat{c}_{k(p)}^+/\hat{c}_{k(p)}$ are the creation/annihilation operators for the electrons in the leads.

Isolated molecule part $\hat{H}_0$ and tunneling part $\hat{H}_{\text{tun}}$ contain electron-phonon interaction, which can be consistently derived by means of the adiabatic approach in the following way. The energies of the molecule electron states $\varepsilon_i(R)$ are functions of interatomic distance R. We would like to mention that the proposed approach could be applied for more complicated molecules. In this case R will correspond to the generalized coordinate of a given vibrational mode. Let us assume that the tunneling process changes occupation of only one electron state ε_1. The Hamiltonian of an isolated molecule $\hat{H}_0$ (including ion motion) in the mixed representation takes the form (hereinafter we assume $\hbar = 1$) [37]

$$\hat{H}_0 = -\frac{1}{2M}\frac{\partial^2}{\partial R^2} + \varepsilon_1(R)\hat{a}_1^+\hat{a}_1 + W_0(R), \tag{3}$$

where $\hat{a}_1$ is the electron annihilation operator for the localized molecule state with the energy ε_1 and $W_0(R)$ is the effective ion interaction energy, corresponding to the empty electron state ε_1. Interaction between ions changes for the occupied electron state ε_1 and it reads

$$W_1(R) = W_0(R) + \varepsilon_1(R). \tag{4}$$

Ion equilibrium positions and vibrational frequencies are different for occupied and empty states of the molecule as they are determined by different potentials. For small molecules with a small number of electrons variation in the occupation of even a single electron state may substantially affect both the nucleus equilibrium positions and vibrational frequencies. For the occupied (empty) state ε_1, the equilibrium position $R_{1(0)}$ is determined by the condition $\frac{\partial}{\partial R}W_{1(0)}(R)|_{R_{1(0)}} = 0$.

One can introduce the creation and annihilation operators $\hat{b}^+$ and $\hat{b}$ to describe nuclei motion in the harmonic approximation in the potential W_0, corresponding to the state with unoccupied level ε_1:

$$(R - R_0) = \sqrt{\frac{1}{2M\omega_0}}(\hat{b} + \hat{b}^+),$$
$$\frac{\partial}{\partial R} = \sqrt{\frac{M\omega_0}{2}}(\hat{b} - \hat{b}^+), \tag{5}$$

where $M\omega_0^2 = \frac{\partial^2}{\partial R^2}W_{(0)}(R)|_{R_{(0)}}$.

To analyze an oscillatory motion of atoms in the potential W_1 with occupied level ε_1 one should use new operators $\hat{b}_1^+$ and $\hat{b}_1$:

$$(R - R_1) = \sqrt{\frac{1}{2M\omega_1}}(\hat{b}_1 + \hat{b}_1^+),$$
$$\frac{\partial}{\partial R} = \sqrt{\frac{M\omega_1}{2}}(\hat{b}_1 - \hat{b}_1^+), \tag{6}$$

with $M\omega_1^2 = \frac{\partial^2}{\partial R^2}W_{(1)}(R)|_{R_{(1)}}$.

To define the uniform operators for creation/annihilation of the vibrational quanta in both cases simultaneously one should introduce the canonical transformation of the $\hat{b}^+/\hat{b}$ operators by means of the unitary operator $\hat{S}$:

$$\hat{\hat{b}} = \hat{S}\hat{b}\hat{S}^{-1},$$
$$\hat{\hat{b}}^+ = \hat{S}\hat{b}^+\hat{S}^{-1}, \tag{7}$$

where

$$\hat{S} = \hat{S}_2\hat{S}_1 \tag{8}$$

and

$$\hat{S}_1 = e^{\frac{\vartheta}{2}\hat{a}_1^+\hat{a}_1(\hat{b}\hat{b} - \hat{b}^+\hat{b}^+)},$$
$$\hat{S}_2 = e^{\lambda\hat{a}_1^+\hat{a}_1(\hat{b} - \hat{b}^+)}. \tag{9}$$

Operator $\hat{S}_1$ is the squeeze operator and $\hat{S}_2$ is the displacement operator. The new creation/annihilation operators $\hat{\hat{b}}^+/\hat{\hat{b}}$ (7) coincide with operators $\hat{b}^+/\hat{b}$ while acting on the empty state with $n_1 = 0$ and with operators $\hat{b}_1^+/\hat{b}_1$ in the case of action on the occupied state with $n_1 = 1$. The parameters λ and ϑ should be chosen as follows:

$$\lambda = \sqrt{\frac{M\omega_0}{2}}(R_0 - R_1), \quad \vartheta = \frac{1}{2}\ln\left(\frac{\omega_1}{\omega_0}\right). \tag{10}$$

Electron operators should be transformed simultaneously by means of the same operator $\hat{S}$:

$$\hat{\hat{a}}_1 = \hat{S}\hat{a}_1\hat{S}^{-1}. \tag{11}$$

Since operator $\hat{n}_1 = \hat{a}_1^+\hat{a}_1$ commutes with the transformation operator $\hat{S}$, the following combination remains invariant: $\hat{\hat{a}}_1^+\hat{\hat{a}}_1 = \hat{a}_1^+\hat{a}_1$. Thus, the electron operators in the initial Hamiltonian (3) can be rewritten in terms of the new operators $\hat{\hat{a}}_1^+\hat{\hat{a}}_1$. Nucleus vibrational motion is described by operators $\hat{\hat{b}}(\hat{\hat{b}}^+)$ for the occupied and empty electron states on equal footing. Consequently, the Hamiltonian of an isolated molecule in the second quantization representation can be written in the form [37]

$$\hat{H}_0 = \tilde{\varepsilon}_1\hat{\hat{a}}_1^+\hat{\hat{a}}_1 + \omega_0\hat{\hat{b}}^+\hat{\hat{b}} + (\omega_1 - \omega_0)\hat{\hat{a}}_1^+\hat{\hat{a}}_1\hat{\hat{b}}^+\hat{\hat{b}}, \tag{12}$$

where the energy of the electron state, $\tilde{\varepsilon}_1 = W_1(R_1) - W_0(R_0) + \frac{1}{2}(\omega_1 - \omega_0)$, does not coincide in the general case with $\varepsilon_1(R_1)$, where the constant contribution $[W_0(R_0) + \frac{1}{2}\omega_0]$ was omitted. Transformed operators' representation has a clear physical meaning. Whereas operator $\hat{b}^+$ is responsible for the transition to the next excited state in the initial oscillator, operator $\hat{\tilde{b}}^+$ describes transition to the next excited state even if the equilibrium position and frequency of the oscillator are modified. This is a "correct" operator which determines the excitation of a molecule vibrational subsystem in any case.

After canonical transformation given by Eqs. (7) and (11) the tunneling Hamiltonian includes a part corresponding to the electron-phonon interaction during the tunneling processes (see [37]). It does not play an important role in the problem of phonon mode squeezing due to the tunneling processes, so hereafter we will consider the tunneling Hamiltonian in a standard form. Tunneling Hamiltonian $\hat{H}_{\text{tun}}$ corresponds to the electron transfer between the leads through the molecule state:

$$\hat{H}_{\text{tun}} = \sum_k t_k(\hat{c}_k^+\hat{a}_1 + \hat{a}_1^+\hat{c}_k) + \sum_p t_p(\hat{c}_p^+\hat{a}_1 + \hat{a}_1^+\hat{c}_p), \quad (13)$$

where $t_{k(p)}$ is the tunneling transfer amplitude between the molecule localized state and the tunneling contact leads, which is considered to be independent of momentum and spin. The tunneling matrix element corresponds to the position of atoms at the distance $(\frac{R_1+R_0}{2})$: $t_{k(p)} = t_{k(p)}(\frac{R_1+R_0}{2})$. We neglect here processes of direct tunneling between the contact leads assuming that they have much smaller amplitude in comparison with tunneling through the molecule states.

Let us calculate explicitly the square displacement of the molecule atoms for the state with the tunneling current. In explicit form, transformation (7) reads

$$\begin{aligned}\hat{\tilde{b}} &= \hat{U}\hat{b} + \hat{V}\hat{b}^+ + \hat{\tilde{b}}_0,\\ \hat{\tilde{b}}^+ &= \hat{U}\hat{b}^+ + \hat{V}\hat{b} + \hat{\tilde{b}}_0,\end{aligned} \quad (14)$$

where

$$\begin{aligned}\hat{U} &= (1-\hat{n}_1) + \hat{n}_1\text{ch}(\vartheta),\\ \hat{V} &= \hat{n}_1\text{sh}(\vartheta),\end{aligned} \quad (15)$$

$\hat{\tilde{b}}_0 = \sqrt{\frac{\omega_1}{\omega_0}}\lambda\hat{n}_1$, and $\hat{n}_1 = \hat{a}_1^+\hat{a}_1$.

From Eq. (14) one can get the following relations for $\hat{\beta} = \hat{\tilde{b}} - \hat{\tilde{b}}_0$:

$$(\hat{\beta} \pm \hat{\beta}^+)^2 = (\hat{U} \pm \hat{V})^2 \cdot (b \pm b^+)^2. \quad (16)$$

Taking into account that

$$\begin{aligned}\hat{U}^2 &= 1 - \hat{n}_1 + \hat{n}_1\text{ch}^2\vartheta,\\ \hat{V}^2 &= \hat{n}_1\text{sh}^2\vartheta,\end{aligned} \quad (17)$$

one can obtain

$$\begin{aligned}(\hat{U} \pm \hat{V})^2 &= [(1-\hat{n}_1) + \hat{n}_1(\text{ch}\vartheta \pm \text{sh}\vartheta)]^2\\ &= 1 - \hat{n}_1 + \hat{n}_1 e^{\pm 2\vartheta} = 1 - \hat{n}_1 \cdot (1 - e^{\pm 2\vartheta}).\end{aligned} \quad (18)$$

The mean-square values of two quadratures $\langle(\hat{\beta} \pm \hat{\beta}^+)^2\rangle$ are connected by the relation

$$\begin{aligned}\langle(\hat{\beta} \pm \hat{\beta}^+)^2\rangle = [1 - \langle n_1\rangle \cdot (1 - e^{\pm 2\vartheta})]\\ \times \langle(\tilde{b} \pm \tilde{b}^+)^2\rangle.\end{aligned} \quad (19)$$

Substituting $\vartheta = (1/2)\ln(\frac{\omega_1}{\omega_0})$ and considering that in the absence of an electron the tunneling molecule is in the equilibrium state $[\langle(\tilde{b} \pm \tilde{b}^+)^2\rangle = 1]$ one can obtain

$$\langle(\hat{\beta} \pm \hat{\beta}^+)^2\rangle = \pm[1 + 2N_B(\omega_0)]\left\{1 - \langle n_1\rangle \cdot \left[1 - \left(\frac{\omega_1}{\omega_0}\right)^{\pm 1}\right]\right\}, \quad (20)$$

since $\langle(\tilde{b} \pm \tilde{b}^+)^2\rangle = \pm[1 + 2N_B(\omega_0)]$, where N_B is the Bose distribution function. In the low-temperature regime the displacement reads [in units $(\hbar/M\omega_0)^{1/2}$]

$$\begin{aligned}\langle(\delta R)^2\rangle &= \frac{1}{2}\langle(\hat{\beta} + \hat{\beta}^+)^2\rangle\\ &= \frac{1}{2}\left\{1 - \langle n_1\rangle \cdot \left[1 - \left(\frac{\omega_1}{\omega_0}\right)^{\pm 1}\right]\right\}.\end{aligned} \quad (21)$$

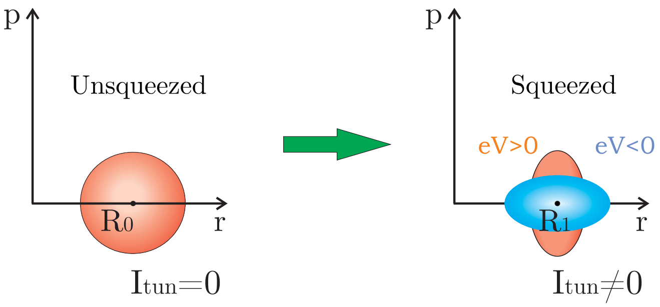

FIG. 2. Sketch of the molecule vibrational mode states squeezing in the presence of tunneling current.

Equation (21) demonstrates the possibility for the molecule vibrational mode state squeezing (see Fig. 2).

If in the absence of the tunneling current electron state ε_1 is empty, then the molecule vibrational mode is described by the operators $\hat{b}$ and $\hat{b}^+$, with the usual ground state $|0\rangle_b$:

$$b|0\rangle_b = 0. \quad (22)$$

But when the electron state ε_1 becomes occupied due to the tunneling current then the new molecule vibrational mode operators $\hat{\tilde{b}}$ and $\hat{\tilde{b}}^+$ act on a squeezed ground state $|0\rangle_{\tilde{b}}$ determined as $|0\rangle_{\tilde{b}} = \hat{S}_1|0\rangle_b$:

$$\tilde{b}|0\rangle_{\tilde{b}} = 0. \quad (23)$$

So, in the presence of the tunneling current the ground state of the molecule vibrational mode becomes squeezed (see Fig. 2). One can see from Eq. (21) that the effect of vibrational frequencies changing is responsible for such squeezing. Thus the modulation of the interatomic potential curvature is the basis of the discussed effect.

When the tunneling current flows through the molecule, nonequilibrium occupation numbers $\langle n_1\rangle$ can be easily found by means of the Keldysh diagram technique. The electron occupation number for elastic tunneling is directly connected with the lesser Keldysh Green's function $iG_{11}^<(t,t) = \langle n_1\rangle$ and is determined by the well-known expression [38,39]

$$\langle n_1\rangle = \frac{\gamma_k n_k + \gamma_p n_p}{\gamma_k + \gamma_p}, \quad (24)$$

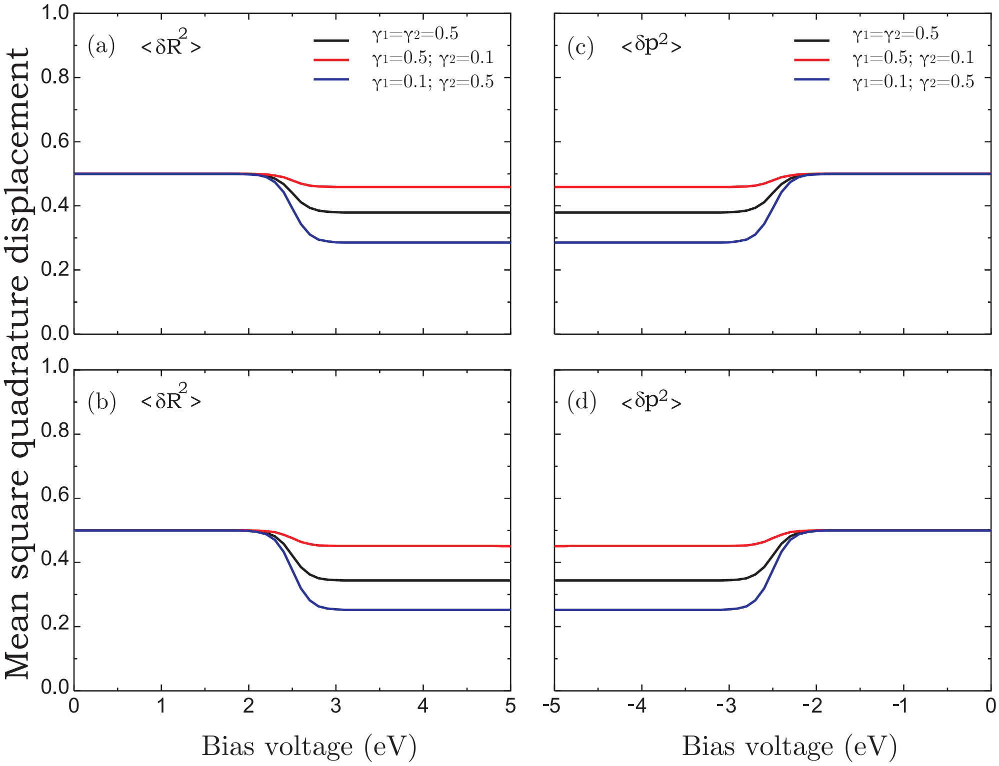

FIG. 3. Molecule quadratures squeezing as a function of applied bias. (a),(c) $\frac{\omega_1}{\omega_0} = 2/3$; $\varepsilon = 2.5$. (b),(d) $\frac{\omega_1}{\omega_0} = 1/2$; $\varepsilon = 2.5$.

where $\gamma_{k(p)} = \pi t^2_{k(p)} \nu^0_{k(p)}$ are the tunneling rates for the electrons transitions between the molecule localized state and the tunneling contact leads. Electron occupation numbers are determined as

$$n_i = \frac{\gamma}{\pi} \cdot \int \frac{f_i(\omega)}{(\omega - \varepsilon_1)^2 + \gamma^2} d\omega, \tag{25}$$

where index $i = k, p$ corresponds to the left (right) lead of the tunneling contact and $f_{k(p)}(\omega)$ is the Fermi distribution function of electrons in the tunneling contact lead. Fermi levels in the k and p leads are shifted by the value of applied bias eV, so $f_k(\omega) = f_F(\omega)$ and $f_p(\omega) = f_F(\omega - \text{eV})$. Relaxation rate γ is the sum of the relaxation rates $\gamma = \gamma_k + \gamma_p$. For asymmetric tunneling contact ($\gamma_k \gg \gamma_p$) the electron occupation number of the localized molecule state $\langle n_1 \rangle$ is close to unity. Depending on the sign of parameter ϑ one of the molecule vibrational mode quadratures has uncertainty less than the fundamental quantum limit.

For small molecules an additional electron can change the effective potential for ions nearly twice. So, the mean-square displacement of the molecule in the presence of tunneling current for $\frac{\omega_1}{\omega_0} \sim 1/2$ reads

$$\langle (\delta R)^2 \rangle \simeq \frac{1}{2} \cdot \left(1 - \frac{\langle n_1 \rangle}{2}\right) \sim \frac{1}{4}. \tag{26}$$

Consequently, the molecule square displacement could be two times smaller than the quantum fundamental limit [see Figs. 3(a) and 3(b)]. So, tunneling current flowing through the molecule electron state results in the squeezing of the vibrational mode state. This effect opens the possibility for molecule localization by the tunneling current. Mean-square displacement suppression depends on the value of parameter $\sqrt{\frac{\omega_1}{\omega_0}}$, which is better pronounced for $\frac{\omega_1}{\omega_0} = 1/2$ in comparison with the situation when $\frac{\omega_1}{\omega_0} = 2/3$ [see Figs. 3(a) and 3(b)]. The same situation occurs for the second quadrature for negative value of applied bias [see Figs. 3(c) and 3(d)]. Now the momentum square displacement reduces in comparison with the fundamental limit.

III. CONCLUSION

We analyzed the influence of the tunneling current on the vibrational mode states of a molecule absorbed on the substrate in the tunneling junction. In the framework of the adiabatic approach two effects were consistently taken into account: changing of equilibrium ion positions and vibrational mode frequency due to modification of the electron occupation number. We revealed that tunneling current flowing through the molecule electron states can result in the squeezing of the vibrational mode state. The effect is due to the changes of the curvature of the interatomic potential and consequently to the modification of the vibrational frequencies due to the electron tunneling to (or from) the molecule. It was demonstrated that the mean-square displacement of the atoms in the molecule and the degree of squeezing could be tuned by the applied bias voltage. This effect opens the possibility to diminish in some sense the zero fluctuations for molecule atoms by the tunneling current.

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