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Tuning of tunneling current noise spectra singularities by localized states charging

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We report the results of theoretical investigations of tunneling current noise spectra in a wide range of applied bias voltage. Localized states of individual impurity atoms play an important role in tunneling current noise formation. It was found that switching "on" and "off" of Coulomb interaction of conduction electrons with two charged localized states results in power law singularity of low-frequency tunneling current noise spectrum ($1/f^{\alpha}$) and also results on high frequency component of tunneling current spectra (singular peaks appear).

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1. Introduction. In the present work we discuss one of the possible reasons for the noise formation in the wide range of applied bias in the STM/STS junctions. Up to now the physical nature and the microscopic origin of tunneling current noise spectra formation in low and high frequency region is in general unknown. Only the limited number of works was devoted to the problem of $1/f^{\alpha}$ noise study and we have found only a few works where high frequency region of the tunneling current spectra is studied [1, 2].

We suggest the theoretical model for tunneling current noise spectra above the flat surface and above the impurity atoms on semiconductor or metallic surfaces. Our model gives an opportunity to describe not only singularities in a low frequency part of tunneling current spectra but also to reveal singular behavior of tunneling current spectra in a high frequency region and to describe spectra peculiarities in a wide range of applied bias voltage. We found experimentally that changing of tunneling current noise spectra above the flat clean surface and above the impurity atom depends on the parameters of tunneling junction such as tip-sample separation or applied bias voltage [3]. Our theoretical model is rather simple, more complicated models describing noise in semiconductors can be found in [4 – 6].

In our previous work we have found that for the low frequency region sudden switching on and off of additional Coulomb interaction in tunneling junction area leads to typical power law dependence of tunneling current noise spectra at the threshold voltage [7]. In this article our aim is to study one of the possible microscopic origins of $1/f^{\alpha}$ noise in tunneling contact in the non-resonance cases and to study high frequency peculiarities of tunneling current spectra in a wide range of applied bias voltage. We shall analize modification of tunneling current noise spectrum by the Coulomb interaction of conduction electrons in the leads (metallic tip and surface) with non-equilibrium localized charges in tunneling contact. It will be shown that corrections to the tunneling vertexes caused by the Coulomb interaction switching on and off result in nontrivial behavior of tunneling current noise spectrum in a wide range of applied bias voltage and should be taken in account.

2. The suggested model and main results. We shall analyze model of two localized states in tunneling contact. In this case one of the localized states is formed by the impurity atom in semiconductor and the other one by the tip apex.

When electron tunnels to or from localized state, the electron filling numbers of localized state rapidly change leading to appearance of localized state additional charge and sudden switching "on" and "off" Coulomb interaction. Electrons in the leads feel this Coulomb potential.

The model system (Fig.1) can be described by hamiltonian $\hat{H}$:

$$\hat{H} = \hat{H}_0 + \hat{H}_{\text{tun}} + \hat{H}_{\text{int}},$$

$$\hat{H}_0 = \sum_p (\varepsilon_p - eV) c_p^+ c_p + \sum_k \varepsilon_k c_k^+ c_k + \sum_{i=1,2} \varepsilon_i a_i^+ a_i,$$
$$\hat{H}_{\text{tun}} = \sum_{k,i} T_{ki} c_k^+ a_i + \sum_{p,i} T_{pi} c_p^+ a_i + T \sum a_1^+ a_2 + \text{h.c.},$$
$$\hat{H}_{\text{int}} = \sum_{k,k'} W_1 c_k^+ c_{k'} a_1 a_1^+ + W_2 c_k^+ c_{k'} a_2 a_2^+.$$

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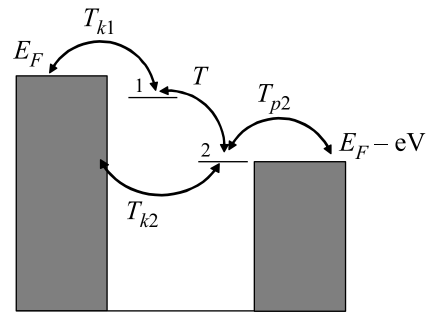

Fig.1. Schematic diagram of tunneling processes through the states localized on impurity atom and on the STM tip apex

$\widehat{H}_0$ describes free electrons in the leads and in the localized states. $\widehat{H}_{tun}$ describes tunneling transitions between the leads through localized states. $\widehat{H}_{int}$ corresponds to the processes of intraband scattering caused by Coulomb potentials W_1, W_2 of localized states charges.

Operators $c_k^+(c_k)$ and $c_p^+(c_p)$ correspond to electrons in the leads and operators $a_i^+(a_i)$ correspond to electrons in the localized states with energy ε_i.

Current noise correlation function is determined as:

$$\begin{aligned}(\hbar/e)^2\cdot S(t,t') &= \langle I_L(t)\cdot I_L(t')\rangle = \\ &= \sum_{k,k',i,j} T_k^2\langle c_k^+(t')a_i(t')a_j^+(t)c_{k'}^+(t)\rangle.\end{aligned}$$

The current noise spectra is determined by Fourier transformation of $S(t,t')$: $S(\omega)=\int S(\tau)d\tau\cdot e^{i\omega\tau}$. We shall use Keldysh diagram technique in our study of low frequency tunneling current noise spectra [8].

Let's consider that STM tip is a metal with a cluster on the tip apex. In this case tip apex localized state energy coincides with tip Fermi level and consequently $eV=\varepsilon_2$.

In our case of weak interaction between localized states ($T<\gamma_{k1},\gamma_{k2},\gamma_{p2}$) possible variants for localized states energy levels position in tunneling contact are: 1. impurity atom localized state energy level exceeds tip apex localized state energy level; 2. tip apex localized state energy level exceeds impurity atom localized state energy level.

Expression which describes tunneling current noise spectra without Coulomb re-normalization can be calculated with the help of Keldysh diagram technic [7, 8]. It consists of three parts.

$$\widetilde{S}_0(\omega)=\widetilde{S}_{01}(\omega)+\widetilde{S}_{02}(\omega)+\widetilde{S}_{03}(\omega).$$

$\widetilde{S}_{01}(\omega)$ and $\widetilde{S}_{02}(\omega)$ are rather simple parts and they have the form:

$$\begin{aligned}(\hbar/e)^2\cdot S_{0i}(\omega) &= \gamma_{ki}^2\int d\omega'\,\mathrm{Im}\,G_{ii}^R(\omega')\times \\ &\times \mathrm{Im}\,G_{ii}^R(\omega+\omega')(n_i(\omega+\omega')-1)\times \\ &\times(n_i(\omega')-n_k(\omega'))+n_i(\omega')\cdot(n_i(\omega+\omega')- \\ &-n_k(\omega+\omega')+\gamma_{ki}^2\int d\omega'\,\mathrm{Im}\,G_{ii}^R(\omega')\times \\ &\times\mathrm{Im}\,G_{ii}^R(\omega+\omega')(n_k(\omega+\omega')-1)n_i(\omega')- \\ &-n_i(\omega')(n_i(\omega+\omega')-1)-n_k(\omega+\omega')n_k(\omega')+ \\ &+n_k(\omega')(n_i(\omega+\omega')+\gamma_{ki}\int d\omega'\,\mathrm{Im}\,G_{ii}^R(\omega+\omega')\times \\ &\times(n_k(\omega'))(n_i(\omega+\omega')-1)+\mathrm{Im}\,G_{ii}^R(\omega')\times \\ &\times(n_i(\omega'))(n_k(\omega+\omega')-1)=\widetilde{S}_{0i},\end{aligned}$$

where $i=1$ corresponds to the $\widetilde{S}_{01}(\omega)$ and $i=2$ corresponds to the $\widetilde{S}_{02}(\omega)$. Green functions shown on the graphs are found in [10]. The contribution of $\widetilde{S}_{01}(\omega)$ is given by graphs with $i=j=1$ (Fig.2a), $\widetilde{S}_{02}(\omega)$ is

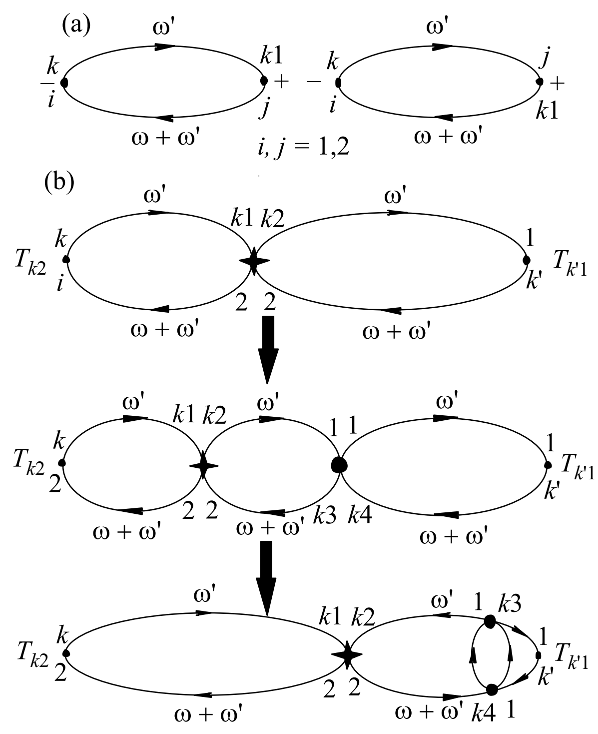

Fig.2. Lowest order diagrams contributing to tunneling current noise spectra for two localized states in tunneling contact. (a) In the absence of Coulomb re-normalization of tunneling vertexes. (b) In the presence of Coulomb re-normalization of tunneling vertexes. Tunneling vertexes are marked by the dot. Coulomb energy W_1 is marked by the bold dot. Coulomb energy W_2 is marked by the star. Solid lines correspond to electron Green functions

described by graph with $i=j=2$ (Fig.2a), $\widetilde{S}_{03}(\omega)$ is given by diagrams with $i\neq j$ (Fig.2a).

$\widetilde{S}_{03}(\omega)$ is not trivial, it exists only due to electron tunneling transitions from one lead to both localized states.

$$
\begin{aligned}
(\hbar/e)^2 S_{03}(\omega) &= 8\gamma_{k_1}\gamma_{k_2}\int d\omega' \mathrm{Im}\, G_{11}^R(\omega') \times \\
&\times \mathrm{Im}\, G_{22}^R(\omega+\omega')(n_1(\omega')(n_2(\omega+\omega')-1) + \\
&+ n_k(\omega')(n_2(\omega+\omega')-1) + (n_1(\omega') \times \\
&\times (n_k(\omega+\omega')-1) - n_k(\omega')(n_k(\omega+\omega')-1) + \\
&+ 8\gamma_{k_1}\gamma_{k_2}\int d\omega' \mathrm{Im}\, G_{22}^R(\omega')\mathrm{Im}\, G_{11}^R(\omega+\omega') \times \\
&\times (n_2(\omega')(n_1(\omega+\omega')-1) + \\
&+ n_k(\omega')(n_1(\omega+\omega')-1) + (n_2(\omega') \times \\
&\times (n_k(\omega+\omega')-1) - n_k(\omega')\cdot(n_k(\omega+\omega')-1) = \widetilde{S}_{03}.
\end{aligned}
$$

In our model relaxation rates γ_{ki}, γ_{pi}, non-equilibrium localized states filling numbers n_1, n_2 and Green functions $G_{ij}^{<}$ are determined from Dyson equations in [7] and [10].

Some typical low frequency tunneling current noise spectra for different values of dimensionless kinetic parameters without Coulomb re normalization are shown on (Fig.3a). It is clearly evident that when frequency aspire to zero tunneling current spectra aspire to constant value for different dimensionless kinetic parameters.

Now let us consider re-normalization of the tunneling amplitude and vertex corrections to the tunneling current spectra caused by Coulomb interaction between both localized states and tunneling contact leads. Renormalization gives us two types of diagrams contributing to the final tunneling current noise spectra expression (Fig.2b). Ladder diagrams is the most simple type of diagrams which gives logarithmic corrections to vertexes. But this is not the only relevant kind of graphs. We must consider one more type of graphs (parquet graphs) which also gives logarithmicaly large contribution to tunneling spectra. In parquet graphs a new type of "bubble" appears instead of "dots" in ladder diagrams (Fig.2b) [7, 9].

The final expression for tunneling current noise spectra after Coulomb re-normalization of tunneling vertexes can be written as:

$$
\begin{aligned}
(\hbar/e)^2\cdot S(\omega) &= \widetilde{S}_0(\omega) + \widetilde{S}_{01}(\omega) \times \\
&\times \left(\left(\frac{D^2}{(\omega+eV+E_1)^2+\Gamma_2^2}\right)^{W_1\nu} + \right. \\
&+ \left.\left(\frac{D^2}{(\omega+eV+E_2)^2+\Gamma_1^2}\right)^{W_1\nu}\right) + \\
&+ \widetilde{S}_{02}(\omega)\cdot\left(\left(\frac{D^2}{(-\omega+eV+E_1)^2+\Gamma_1^2}\right)^{W_2\nu} + \right. \\
&+ \left.\left(\frac{D^2}{(-\omega+eV+E_2)^2+\Gamma_2^2}\right)^{W_2\nu}\right) + \widetilde{S}_{03}(\omega) \times \\
&\times \left(\left(\frac{D^2}{(\omega+eV+E_1)^2+\Gamma_2^2}\times\right)^{W_1\nu} + \right. \\
&+ \left.\left(\frac{D^2}{(\omega+eV+E_2)^2+\Gamma_1^2}\right)^{W_1\nu}\right) \times \\
&\times \left(\left(\frac{D^2}{(-\omega+eV+E_1)^2+\Gamma_1^2}\right)^{W_2\nu} + \right. \\
&+ \left.\left(\frac{D^2}{(-\omega+eV+E_2)^2+\Gamma_2^2}\right)^{W_2\nu}\right),
\end{aligned}
$$

where

$$
\begin{aligned}
E_{1,2} &= -\frac{\varepsilon_1+\varepsilon_2}{2} \pm \frac{\sqrt{(\varepsilon_1-\varepsilon_2)^2+4T^2}}{2}, \\
\Gamma_1 &\sim \Gamma_2 \sim (\gamma_{k2}+\gamma_{p2}+\gamma_{k1}),
\end{aligned}
$$

where D – are the bandwidths of right and left leads, ν – the equilibrium density of states in the tunneling contact leads, W — Coulomb energy.

In the case of strong interaction between localized states in tunneling contact we have localized states energy levels splitting and as a result there is no singularity in tunneling current spectra at the low frequency region. In our situation of weak interaction between localized states ($T < \gamma_{k1}, \gamma_{k2}, \gamma_{p2}$) tunneling transfer amplitude T plays an important role in kinetic processes in the tunneling junction but weakly influences on the energy spectra.

First of all we consider the situation when both localized states acquire positive charge. Fig.3b demonstrate tunneling current spectra for typical values of dimensionless kinetic parameters. We can see that renormalization of tunneling matrix element by switched "on" and "off" Coulomb interaction of charged impurities leads to typical power law singularity in low frequency part of tunneling current noise spectra and to the peak in the particular high frequency region, caused by the singularity on the frequency $\omega = \varepsilon_2 - \varepsilon_1$.

Tunneling current noise spectra in double logarithmic scale demonstrate frequency regions where every part of final expression which include different power law exponent, approximate noise spectra in the best way Fig.3c.

Let's analyze tunneling current spectra shown on Fig.3c–e. In the case of two interacting positively charged localized states the tunneling current spectra at low frequency and in the region of the high frequency singularity is always determined by the term which produces the most strong of logarithmic singularity, determined by the sum of localized states Coulomb energies

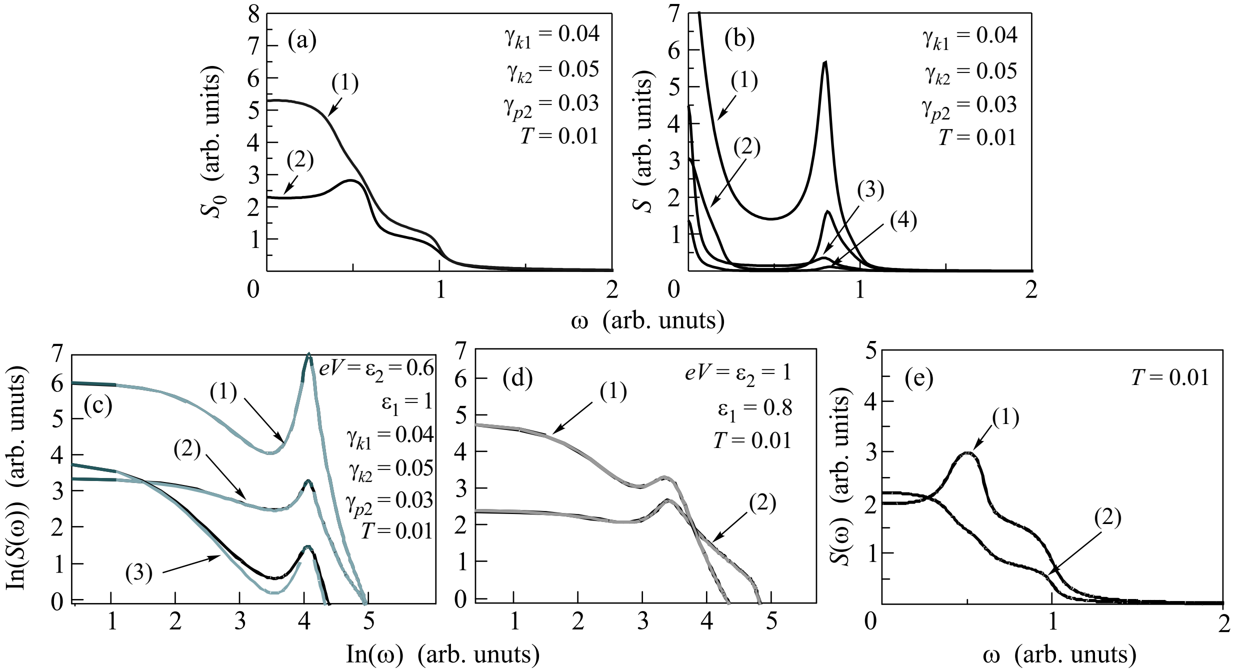

Fig.3. Typical tunneling current noise spectra for different values of dimensionless kinetic parameters for two localized states in tunneling contact ($eV = \varepsilon_2 \neq \varepsilon_1$). a) In the absence of Coulomb re-normalization of tunneling vertexes (1) $eV = \varepsilon_2 = 1$, $\varepsilon_1 = 0.6$; (2) $eV = \varepsilon_2 = 0.6$, $\varepsilon_1 = 1$. b) In the presence of Coulomb re-normalization of tunneling vertexes (1) $eV = \varepsilon_2 = 1$, $\varepsilon_1 = 0.2$, $W_1\nu = 0.4$, $W_2\nu = 0.3$; (2) $eV = \varepsilon_2 = 0.2$, $\varepsilon_1 = 1$, $W_1\nu = 0.4, W_2\nu = 0.3$; (3) $eV = \varepsilon_2 = 1$, $\varepsilon_1 = 0.2$, $W_1\nu = 0,6$, $W_2\nu = 0.1$; (4) $eV = \varepsilon_2 = 0.2$, $\varepsilon_1 = 1$, $W_1\nu = 0.6$, $W_2\nu = 0.1$. c) In the presence of Coulomb re-normalization of tunneling vertexes in double logarithmic scale when both localized states acquire positive charge (1) $W_1\nu = 0.1$, $W_2\nu = 0.6$; (2) $W_1\nu = 0.4$, $W_2\nu = 0.3$; (3)$W_1\nu = 0.6$, $W_2\nu = 0.1$. d) In the presence of Coulomb re-normalization of tunneling vertexes in double logarithmic scale when localized states acquire charges of different signs (1) $\gamma_{k1} = 0.05$, $\gamma_{k2} = 0.03$, $\gamma_{p2} = 0.04$, $W_1\nu = 0.6$, $W_2\nu = -0.1$; (2) $\gamma_{k1} = 0.04$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.03$, $W_1\nu = -0.4$, $W_2\nu = 0.3$. e). In the presence of Coulomb re-normalization of tunneling vertexes when both localized states acquire negative charge (1) $eV = \varepsilon_2 = 0.6$, $\varepsilon_1 = 1$, $\gamma_{k1} = 0.04$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.03$, $W_1\nu = -0.4$, $W_2\nu = -0.3$; (2) $eV = \varepsilon_2 = 1$, $\varepsilon_1 = 0.6$, $\gamma_{k1} = 0.05$, $\gamma_{k2} = 0.03$, $\gamma_{p2} = 0.04$, $W_1\nu = -0.4$, $W_2\nu = -0.3$

at any values of dimensionless tunneling rates of tunneling contact. (Fig.3c). If one of the Coulomb energies strongly exceeds the other one this potential determine tunneling current noise spectra with increasing of frequency (Fig.3c).

Tunneling current noise spectra in double logarithmic scale (Fig.3d) make it clear that when localized states acquire charges of opposite signs tunneling current noise spectra in the low frequency region and in the region of the high frequency singularity ($\omega = \varepsilon_2 - \varepsilon_1$) are always approximated by the term depending on maximum positive value of impurity atom Coulomb energy W_1 or tip apex localized state Coulomb energy W_2. Tunneling current noise spectra in double logarithmic scale in the situation when both localized states acquire negative charges (Fig.3e) demonstrate that singular effects become negligible.

Now let's describe interaction effects in tunneling contact when both localized states are formed by impurity atoms in the surface. In this case we can say about cluster in the surface $eV \neq \varepsilon_2 \neq \varepsilon_1$.

Typical low frequency tunneling current noise spectra for different values of dimensionless kinetic parameters without Coulomb re-normalization are shown on Fig.4a.

It is clearly evident that when frequency aspire to zero tunneling current spectra aspire to constant value for different dimensionless kinetic parameters (Fig.4a).

Let us study re-normalized spectra. Fig.4b demonstrate tunneling current noise spectra for typical values of dimensionless kinetic parameters. It is clearly evident that when frequency aspire to zero tunneling current spectra aspire to constant value and we have no power law singularity in a low frequency part of tunneling current spectra.

We can see that re-normalization of tunneling matrix element by switched "on" and "off" Coulomb interaction of charged impurities leads to the singular peaks in high

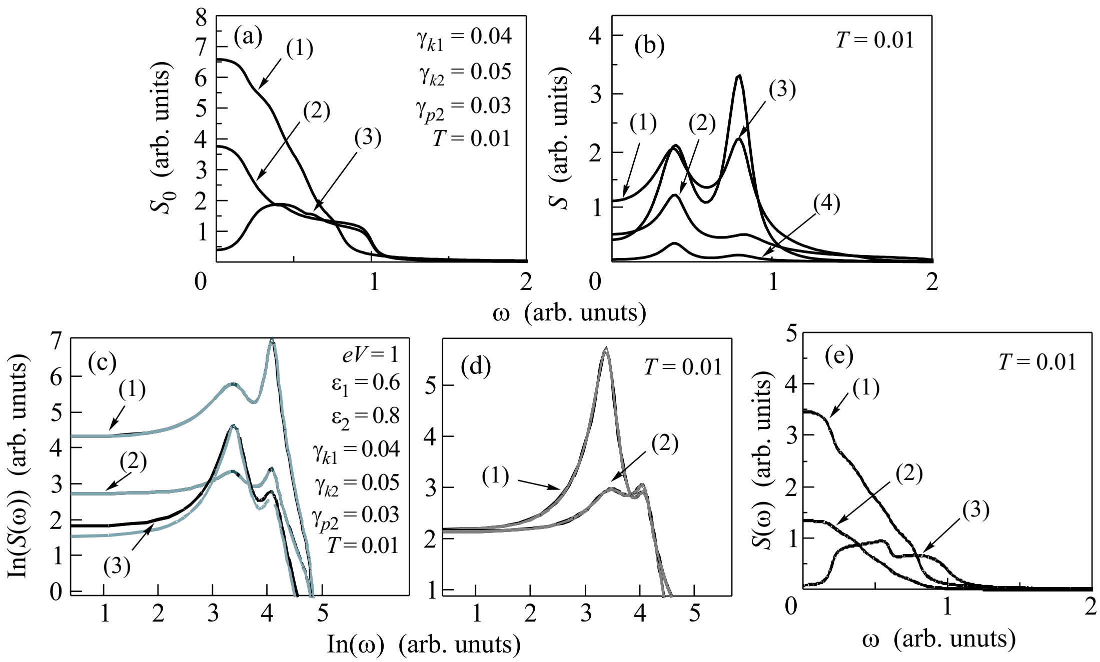

Fig.4. Typical tunneling current noise spectra for different values of dimensionless kinetic parameters for two localized states in tunneling contact ($eV \neq \varepsilon_2 \neq \varepsilon_1$). a) In the absence of Coulomb re-normalization of tunneling vertexes (1) $eV = 1, \varepsilon_1 = 0.6$, $\varepsilon_2 = 0.8$; (2) $eV = 0.6, \varepsilon_1 = 0.4$, $\varepsilon_2 = 1$; (3) $eV = 0.4, \varepsilon_1 = 0.6$, $\varepsilon_2 = 1$. b) In the presence of Coulomb re-normalization of tunneling vertexes (1) $eV = 1, \varepsilon_1 = 0.6$, $\varepsilon_2 = 0.8$, $\gamma_{k1} = 0.04$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.03$, $W_1\nu = 0.4$, $W_2\nu = 0.3$; (2) $eV = 1, \varepsilon_1 = 0.6$, $\varepsilon_2 = 0.8$, $\gamma_{k1} = 0.04$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.03$, $W_1\nu = 0.6$, $W_2\nu = 0.1$; (3) $eV = 0.6, \varepsilon_1 = 0.4$, $\varepsilon_2 = 1$, $\gamma_{k1} = 0.05$, $\gamma_{k2} = 0.04$, $\gamma_{p2} = 0.03$, $W_1\nu = 0.4$, $W_2\nu = 0.3$; (4) $eV = 1, \varepsilon_1 = 0.8$, $\varepsilon_2 = 0.6$, $\gamma_{k1} = 0.03$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.04$, $W_1\nu = 0.6$, $W_2\nu = 0.1$;. c) In the presence of Coulomb re-normalization of tunneling vertexes in double logarithmic scale when both localized states acquire positive charge (1) $W_1\nu = 0.1$,$W_2\nu = 0.6$; (2) $W_1\nu = 0.4$, $W_2\nu = 0.3$; (3) $W_1\nu = 0.6$, $W_2\nu = 0.1$. d)In the presence of Coulomb re-normalization of tunneling vertexes in double logarithmic scale when localized states acquire charges of different signs (1) $eV = 1, \varepsilon_1 = 0.6$, $\varepsilon_2 = 0.8$, $\gamma_{k1} = 0.03$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.04$, $W_1\nu = 0.6$, $W_2\nu = -0.1$; (2) $eV = 0.8, \varepsilon_1 = 0.4$, $\varepsilon_2 = 1$, $\gamma_{k1} = 0.05$, $\gamma_{k2} = 0.03$, $\gamma_{p2} = 0.04$, $W_1\nu = -0.4$, $W_2\nu = 0.3$. e)In the presence of Coulomb re-normalization of tunneling vertexes in double logarithmic scale when both localized states acquire negative charge (1) (1) $eV = 1, \varepsilon_1 = 0.6$, $\varepsilon_2 = 0.8$, $\gamma_{k1} = 0.04$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.03$, $W_1\nu = -0.4$, $W_2\nu = -0.3$; (2) $eV = 1, \varepsilon_1 = 0.8$, $\varepsilon_2 = 0.6$, $\gamma_{k1} = 0.05$, $\gamma_{k2} = 0.04$, $\gamma_{p2} = 0.03$, $W_1\nu = -0.6$, $W_2\nu = -0.1$; (3) $eV = 0.4, \varepsilon_1 = 1$, $\varepsilon_2 = 0.8$, $\gamma_{k1} = 0.03$, $\gamma_{k2} = 0.05$, $\gamma_{p2} = 0.04$, $W_1\nu = -0.1$, $W_2\nu = -0.6$

frequency regions of tunneling current spectra, caused by singularities on the frequencies $\omega = eV - \varepsilon_1$ and $\omega = eV - \varepsilon_2$.

Let's analyze tunneling current spectra in double logarithmic scale shown on Fig.4c–e. In the case of two interacting positively charged localized states the tunneling current spectra in the regions of the singular peaks is determined by the term which produces the most strong of logarithmic singularity, determined by the sum of localized states Coulomb potentials at any values of dimensionless tunneling rates of tunneling contact. (Fig.4c). If one of the Coulomb potentials strongly exceeds the other one this potential determine tunneling current noise spectra except frequency regions in the vicinity of singular peaks (Fig.4c).

Tunneling current noise spectra in double logarithmic scale (Fig.4d) make it clear that when localized states acquire charges of opposite signs tunneling current noise spectra is always approximated by the term depending on maximum positive value of impurity atom Coulomb energy W_1 or tip apex localized state Coulomb energy W_2.

Tunneling current noise spectra in double logarithmic scale in the situation when both localized states acquire negative charges (Fig.4e) make it clearly evident that singular effects after Coulomb re-normalization become negligible. Simple estimation gives possibility to analyze the validity of obtained results. For typical composition of tunneling junction parameters in the low frequency region power spectrum of tunneling current corresponds to experimental results. Power spectrum on zero frequency has the form:

$$S(0) \approx (\gamma_{\text{eff}} \cdot e/\hbar)^2 \cdot (D/\gamma_{\text{eff1}})^{\nu \cdot W} \cdot (1/\bigtriangleup \omega)$$

For typical γ_{eff}, $\gamma_{\text{eff1}} \approx 10^{-13}$, $D \approx 10$, $W \approx 0.5$, $S(0) \approx 10^{-18}\,\text{A}^2/\text{Hz}$ [3].

3. Conclusion. The microscopic theoretical approach describing tunneling current noise spectra in a wide range of applied bias voltage taking in account many-particle interaction was proposed.

In the case of two localized states in tunneling junction when energy level of one of the localized states is connected with the tip apex and is not equal to energy level of the impurity atom localized state on the surface we can see typical power law dependence for the low frequency part of tunneling current spectra and singular peak in the particular high frequency region of tunneling current spectra determined by the localized states energy levels deposition in the tunneling junction ($eV = \varepsilon_2 \neq \varepsilon_1$). In the non-resonance case of the cluster on the surface ($eV \neq \varepsilon_2 \neq \varepsilon_1$) we have found two singular peaks in the high frequency region of tunneling current noise spectra. So our results demonstrate that changing of the applied bias voltage leads to the tuning of tunneling current noise spectra.

This effect can be qualitatively understood by the following way: apart from the direct tunneling from the localized state to the state with momentum k in the lead of tunneling contact described by the amplitude T_k, the electron can first tunnel into any other empty state k' in the lead and then scatter to the state k by Coulomb potential. So the appearance of even a weak Coulomb interaction significantly modifies the many particle wave function of the electron gas in the leads.

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