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Coulomb correlation effects in tunneling processes through localized impurity states *

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Abstract

Non-equilibrium Coulomb effects in resonant tunneling through deep impurity states are analyzed. It is shown that corrections to the tunneling vertex caused by the Coulomb potential result in a nontrivial behavior of tunneling characteristics and should be taken into account. One encounters effects similar to the Mahan edge singularities in the problem of X - ray absorption spectra in metals. The Coulomb vertex corrections lead to a smoothed power law singularity in current-voltage characteristics. The effect is well pronounced if tunneling rate from a deep impurity level to metallic tip is much larger than relaxation rate of non-equilibrium electron density at the localized state. This condition can be satisfied in experiments with a deep impurity state in the semiconductor gap.

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Coulomb interaction plays great role in tunneling processes in ultra small junctions. Experimentally investigated junctions are of very different nature, made by different technology, so in various experiments various Coulomb effects can be observed. First of all Coulomb blockade and Kondo effects

*Presented at Russian-Israeli Conference *Frontiers in Condensed Matter Physics.* Shoresh, Israel, 19-24 October 2003 should be mentioned here. Now it is clear that non-equilibrium electron distribution in the tunneling contact area and interaction between tunneling particles give rise to strong modification of initial local density of states and tunneling conductivity spectra. We show that tunneling transfer amplitude itself can be also changed by Coulomb interaction of conduction electrons in a lead with charges localized at impurity states in contact area. If the renormalization of tunneling amplitude is strong enough then smoothed power-law singularity in current-voltage characteristics appears near some threshold voltage.

Let us discuss the junction of the type: semiconductor-impurity state-metallic lead. This system can be described by the Hamiltonian $\hat{H}$:

$$\hat{H} = \hat{H}_L + \hat{H}_R + \hat{H}_{imp} + \hat{H}_T + \hat{H}_{int}, \tag{1}$$

where:

$$\hat{H}_R = \sum_{k\sigma} (\varepsilon_k - \mu) c_{k\sigma}^{+} c_{k\sigma}, \hat{H}_L = \sum_{p\sigma} (\varepsilon_p - \mu - eV) c_{p\sigma}^{+} c_{p\sigma} \tag{2}$$

describes the electron states in the metallic lead (tip) and the semiconductor correspondingly, $c_{k\sigma}^{+}(c_{k\sigma})$ and $c_{p\sigma}^{+}(c_{p\sigma})$ describe creation (annihilation) of electron in states $(k\sigma)$ and $(p\sigma)$ in metal and semiconductor.

$$\hat{H}_{imp} = \sum_{d\sigma} \varepsilon_d c_{d\sigma}^{+} c_{d\sigma} + U n_{d\sigma} n_{d-\sigma} \tag{3}$$

corresponds to a localized impurity state. We shall consider the situation with single occupied impurity level at zero applied voltage due to the on-site Coulomb interaction. However, when analysing behavior of tunnelling current at applied voltage close to the impurity energy ε_d, the on-site Coulomb repulsion of localized electrons can be omitted, because in this situation the impurity state becomes nearly empty above the threshold value of the applied bias. Let us also point out, that the Kondo regime is destroyed on Anderson impurity for the values of applied bias approaching the threshold [1, 2]. In this case the Kondo-effect is not responsible for any unusual features of the tunnelling characteristics .

Tunnelling transitions from the impurity state to the semiconductor and the metal are described by the term:

$$\hat{H}_T = \sum_{kp} (T_{kd} c_{k\sigma}^{+} c_{d\sigma} + T_{pd} c_{p\sigma}^{+} c_{d\sigma}) + h.c.. \tag{4}$$

And, finally, the term H_{int} includes the Coulomb interaction of the core (impurity) hole with conduction electrons in the metal.

$$\hat{H}_{int} = \sum_{kk'\sigma\sigma'} W_{kk'} c^{+}_{k\sigma} c_{k'\sigma}(1 - c^{+}_{d\sigma'} c_{d\sigma'}). \qquad (5)$$

Hamiltonian H_{int} appears as a many-particle interaction and describes rearrangement of conduction electrons in the potential of the hole, suddenly "switched on" by tunnelling transition of the impurity electron. Scattering by the impurity hole Coulomb potential does not change the electron spin. Thus in the lowest order in T_{kd} we can consider renormalization of the tunnelling amplitude independently for each spin - the same one for conduction and impurity electrons. We use for simplicity an averaged value of screened Coulomb interaction describing s - wave scattering of conduction electrons by a deep hole $W_{kk'} = W$. It is convenient to describe the tunneling current in the framework of Keldysh diagram technique for non-equilibrium processes [3]. The general form of kinetic equations is

$$(\widehat{G}_0^{-1} - \widehat{G^*}_0^{-1})\widehat{G}^< = (\widehat{\Sigma}\widehat{G})^< - (\widehat{G}\widehat{\Sigma})^<,$$

where $\widehat{\Sigma}$ usually includes all the interactions. If $\widehat{\Sigma}$ is determined only by the tunneling coupling to the leads, then the kinetic equation takes the form

$$i\frac{\partial}{\partial t} G^<_{dd} = \sum_p T_p (G^<_{pd} - G^<_{dp}) + \sum_k T_k (G^<_{kd} - G^<_{dk}).$$

The right-hand side of this equation determines the currents from impurity to semiconductor and to metal lead respectively. We rewrite the current in the following way (we set charge $e = 1$):

$$I(V) = Im(J(V)), \qquad J(V) = i\sum_{k,\sigma} \int d\omega T_{kd} G^{\sigma<}_{kd}, \qquad (6)$$

where we have defined a tunnelling "response function" $J(V)$. If the Coulomb interaction is neglected one can obtain the usual expression for this response function in the lowest order in T_{kd}:

$$J^0(V) = i\sum_{k,\sigma} \int d\omega T^2_{kd} (G^{\sigma<}_{kk} G^{\sigma A}_{dd} + G^{\sigma R}_{kk} G^{\sigma<}_{dd}) \qquad (7)$$

Substituting the corresponding expressions for the Keldysh functions [4] and performing integration over k we get:

$$J^0(V) = \gamma_t \int d\omega \left[\frac{n_k^0(\omega)}{\omega + eV - \varepsilon_d + i(\gamma + \gamma_t)} + \frac{n_d(\omega)(-i(\gamma + \gamma_t))}{(\omega + eV - \varepsilon_d)^2 + (\gamma + \gamma_t)^2} \right], \tag{8}$$

where the tunnelling rate $\gamma_t = T_{kd}^2 \nu$, and ν the unperturbed density of states in the metallic tip. Kinetic parameter γ corresponds to the relaxation rate of electron distribution in the localized state. In the suggested microscopic picture (eq.(4)) this relaxation rate is determined by weak enough electron tunnelling transitions from the impurity to the semiconductor continuum states $\gamma = T_{pd}^2 \nu_p$. (In general γ can include different types of relaxation processes.)

Non-equilibrium impurity filling numbers $n_d(\omega)$ are determined from kinetic equations for the Keldysh functions $G^<$:

$$n_d(\omega) = \frac{\gamma n_p^0(\omega) + \gamma_t n_k^0(\omega)}{\gamma + \gamma_t}. \tag{9}$$

Thus for low temperatures :

$$\begin{aligned} J^0(V) &= \gamma_t \, ln(|X|) \\ &+ i \frac{\gamma_t \gamma}{\gamma + \gamma_t} \left[\mathrm{arcctg}((eV - \varepsilon_d)/(\gamma + \gamma_t)) - \mathrm{arcctg}((-\varepsilon_d)/(\gamma + \gamma_t)) \right] \end{aligned} \tag{10}$$

with

$$X = (eV - \varepsilon_d + i(\gamma + \gamma_t))/D), \tag{11}$$

where D is the band width for electrons in metal.

The usual form of the tunnelling current is of course reproduced from Eqs.(6, 8, 10).

$$I^0(V) = \frac{\gamma_t \gamma}{\gamma + \gamma_t} \int ImG_{d,d}^R(\omega)(n^0(\omega) - n^0(\omega - eV))d\omega.$$

Now let us consider renormalization of the tunnelling amplitude and vertex corrections to the tunnelling current caused by the Coulomb interaction between the impurity core hole and electrons in the metal. Many particle picture strongly differs from the single-particle one near the threshold voltage. First order corrections due to the Coulomb interaction (the first graph

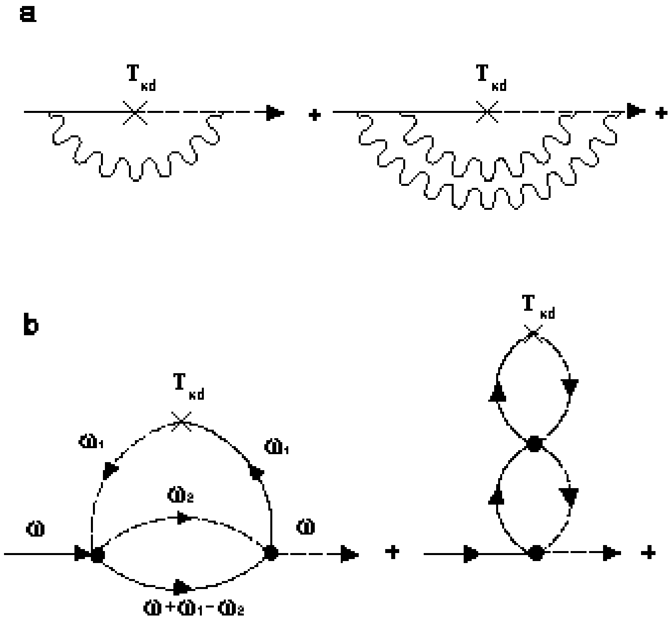

Figure 1: Coulomb corrections to T_{kd}. Solid lines represent G_k and dashed lines — G_d. a)Ladder approximation, b)Parquet graphs (Coulomb wavy lines are redrawn as black circle vertexes).

in Fig. 1a) has a logarithmic divergency at the threshold voltage $eV = \varepsilon_d$, which is cut off by the finite relaxation and tunnelling rates.

$$J^1(V) = i \sum_{k,\sigma} \int d\omega T_{kd}(-G_{kk}^{\sigma<} G_{dd}^{\sigma A} T_{kd}^{1++} + G_{kk}^{\sigma R} G_{dd}^{\sigma<} T_{kd}^{1--}). \qquad (12)$$

Here tunnelling matrix elements are changed in first order by the Coulomb interaction:

$$T_{kd}^{1--} = \sum_{k,\sigma} \int d\omega T_{kd} W (G_{kk}^{\sigma<} G_{dd}^{\sigma A} + G_{kk}^{\sigma R} G_{dd}^{\sigma<}). \qquad (13)$$

We see from Eq. (9) that for $\gamma_t \gg \gamma$ the impurity level becomes nearly empty when the value of applied bias voltage crosses the impurity energy and $n_k^0(\varepsilon_d) \simeq 0$. Though $n_p^0(\varepsilon_d) = 1$, $n_d(\varepsilon_d) << 1$ and there is really a positively charged hole in the impurity state. In this situation only the first term in Eq.(13) is relevant above the threshold voltage. Thus a logarithmic contribution comes from the following combination of the Green functions: $G_{kk}^{\sigma<}G_{dd}^{\sigma A}$. In what follows we retain only logarithmically large parts, assuming that $|ln((\gamma+\gamma_t))/D)| \gg 1$, so only these combinations of Green functions are the most important in perturbation series. Then we obtain from (13) that the tunnelling amplitude contains a logarithmic correction: $T_{kd}^{1--} = -T_{kd}L$, $T_{kd}^{1++} = -T_{kd}^{1--}$, where factor L:

$$L = (W\nu)ln(X). \tag{14}$$

In high orders of perturbation expansion ladder graphs (Fig. 1a) are the simplest "maximally singular" graphs. But this is not the only relevant kind of graphs. If we look at the first graph in Fig. 1b, we notice, that a new type of "bubble" appears, which is logarithmically large for small "total" energy $(\omega+\omega_1)$. The important point is that the relevant region of integration over ω and ω_1 is that of small ω. It is just this region which gives essential contribution to the logarithmic factor L in any other pair of $G_{kk}^{<}G_{dd}^{A(R)}$.

It means that the central bubble also gives an additional logarithmic factor to the total result. In this situation, which is not new in physics, one should retain in the n - th order of perturbation expansion the most divergent terms proportional to $(W\nu)^n L^{n+1}$. The discussed effect is mathematically similar to the Mahan edge singularities in the problem of X - ray absorption spectra in metals [5]. We can say that we deal with a response of Fermi sea to a "sudden switching" of the Coulomb potential when the applied bias crosses the impurity level, though this "sudden switching" develops not in time but in voltage changes. Of course, here we use words "singular" and "divergent" not literally, but only to stress the appearance of large logarithmic factor. The method of summation of these graphs was developed by Dyatlov et al [6]. It was shown that for a proper treatment of this problem one should write down integral equations for so-called parquet graphs (Fig. 1b), which are constructed by successive substitution the simple Coulomb vertex for the two types of bubbles in perturbation series. These equations represent some extension of the ordinary Bethe-Salpeter equation and describe multiple scattering of conduction electrons by the core hole Coulomb potential in the two "most singular" channels. The integral equations can be solved with logarithmic accuracy, as it was done, for example, by Nozieres [7, 8] for edge singularities in X - ray absorption spectra in metals.

Summing up the most divergent graphs with logarithmic accuracy one

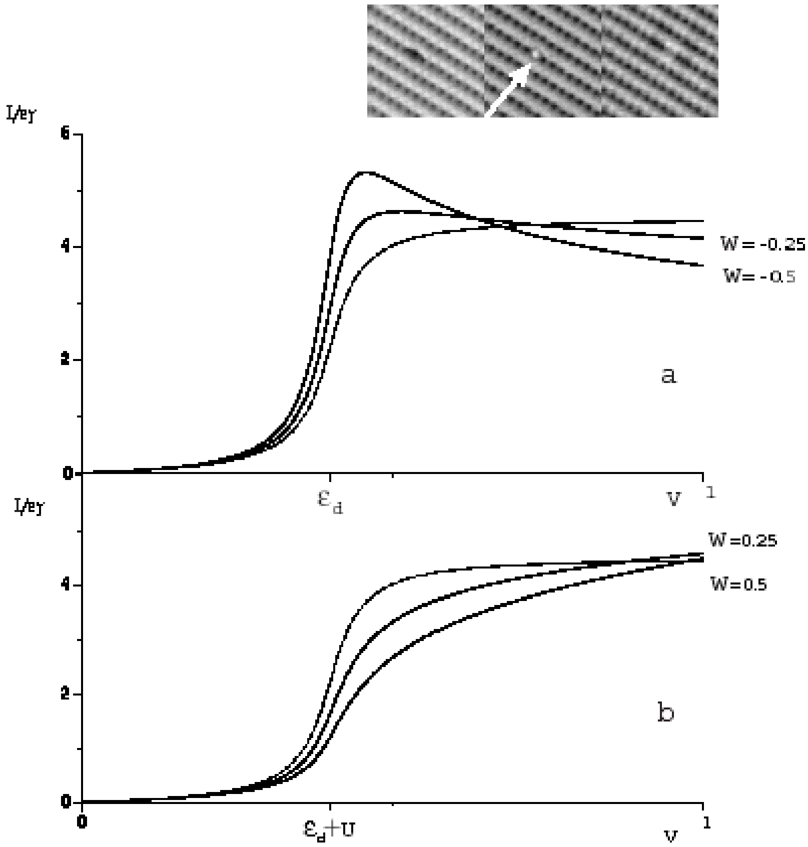

Figure 2: Current-voltage curves for typical values of dimensionless Coulomb and kinetic parameters. a) $w = W\nu < 0$, b) $w = W\nu > 0$, $\gamma_t/\gamma = 3, \varepsilon_d/\gamma = 40$. Dashed line correspond to $W = 0$. Experimental STM image of Cr impurity on InAs (110) surface is shown in the inset for V=0, V=0.5, V=1.5 in sequence.

obtains the following singular part of the response function [7]:

$$J(V) = \frac{\gamma_t(1 - \exp(-2L))}{2W\nu}. \tag{15}$$

Then the tunnelling current near the threshold voltage can be expressed as:

$$I(V) = \frac{\gamma_t}{2W\nu}\left[\frac{D^2}{(eV - \varepsilon_d)^2 + (\gamma_t + \gamma)^2}\right]^{W\nu} \sin\left(2W\nu\phi\right), \tag{16}$$

where $\phi = \text{arcctg}(\frac{eV-\varepsilon_d}{\gamma+\gamma_t})$. If we consider a deep impurity state in the gap of the semiconductor (below the Fermi level) and positive tip bias voltage, then $\varepsilon_d < 0, eV < 0$. So the phase ϕ is a step-like function varying from 0 to π, if the applied bias crosses the threshold $eV = \varepsilon_d$. Since we retain only the most logarithmically large terms in the tunnelling current, Eq. (16) is valid only if $|eV - \varepsilon_d| \ll D$. Eq. (16) shows that the tunnelling current has a peak above the threshold voltage, and then the region of negative differential conductivity begins. In the absence of Coulomb interaction ($W = 0$) this singular part reduces to the usual first order contribution arising from the first term in Eqs. (7, 8).

It seems also possible to set up an experiment with negative impurity charge and negative tip voltage close to the value ε_d+U. In this case $W > 0$, and the Coulomb corrections to the tunnelling amplitude result in power-low behavior of the tunnelling current with the opposite sign of exponent in Eq. (16). The tunnelling current is suppressed near the threshold, compared to the noninteracting case. Current-voltage characteristics obtained for typical values of parameters are shown in Fig. 2.

Experimental STM/STS investigations of deep impurity levels on semiconductor surfaces give evidence of existence of the described effects. Some STM images demonstrate non-monotonic dependence of tunnelling current on the applied bias voltage [9]. Recent STS measurements [10] on Mn doped InAs single crystal surfaces demonstrate I-V characteristics very similar to those discussed in the present paper. Unusual consequence of Eq. (16) with negative exponent in power is, that the current *itself* could be suppressed, but *normalized* tunneling conductivity is enhanced. So the renormalization of the tunneling transfer amplitude by the Coulomb interaction is some new nontrivial effect which can be observed in special experimental setup.

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