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Fast transient current response to switching events in short chains of molecular islands

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Abstract We consider current flow between two metallic leads joined by a short chain of molecular islands interconnected by tunneling junctions. These junctions are assumed to be ”wiggly”, that is switching on and off intermittently. The resulting current response is composed of fast transients which may or may not overlap depending on the ratio between the interplay of the switching times and the dynamical characteristics of the chain, notably the formation time of the quasiparticles and the decay time of ”initial” correlations induced by each switching event. The process is described in terms of non-equilibrium Green’s functions using a combination of partition techniques in space and in time to solve the integrodifferential equations for the particle correlation function numerically. Several model for the switching are explored, among them the case of bursts of swichings within a limited time followed by a quiescent period. For this, we concentrate on the gradual transition to the steady transport regime.

Keywords non-equilibrium · molecular islands · initial correlations · transient currents

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1 Introduction

In this paper, we study numerically transient electronic currents induced by rapid switching in the prototype model of a nanoscopic structure, a single level molecular bridge between two current leads:

The two leads are connected to the central island each by its own independent tunneling junction, left and right. This model has been studied extensively, as reviewed in [1]. The model sophistication has been gradually improved. For a fully systematic account of many-electron interactions based on the modern time-dependent density functional theory see [2]). For inclusion of phonon reservoirs see e.g. [3]. Less attention has been paid to the fast transient currents [4] and [5]. The latter reference provides an extended formal background to the present paper. Most of the work on the molecular bridge model, including ours, has been based quite naturally on the the use of non-equilibrium Green’s functions (NGF). Two fundamental references are [6] for stationary currents and in particular Ref. [7] for time-dependent phenomena, our starting point.

We will make the common simplifications for the electronic Hamiltonian, in particular we follow [6] and [7] and ignore the time dependent capacitance effects at the junctions. The leads will be viewed as reservoirs of independent electrons and also the many body interactions will be ignored within the central island. Altogether, our model will have a purely one-electron character. The emphasis will be on the short time transient dynamic following a rapid switching in a junction and its basic features are properly accounted for by this simple model mimicking well the many body interactions [5].

2 Model and Formalism

The one-electron Hamiltonian for the bridge model is taken in its simplest possible form[5], specializing further the Hamil-

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tonian in [7]:

$$\begin{aligned}\mathscr{H}(t) &= \mathscr{H}_0 + \mathscr{H}'(t)\\ \mathscr{H}_0 &= \mathscr{H}_{0L} + \mathscr{H}_{0B} + \mathscr{H}_{0R}\end{aligned} \tag{1}$$

$$\mathscr{H}'(t) = \alpha_L(t)\mathscr{V}_L + \alpha_R(t)\mathscr{V}_R \tag{2}$$

The specific choice of various parts of the Hamiltonian is as follows. The three parts of the structure, taken as separate, have no time dependence. The leads, labeled by L and R, couple only to the bridge B, but not one to another. The coupling is specified by time independent operator amplitudes $\mathscr{V}_{L,R}$ coupling the leads to the central island orbital. All time dependence in the model is concentrated into two scalar functions $\alpha_{L,R}(t)$. These functions may be arbitrary, but to describe the switching processes, we will assume them to jump suddenly between 0 and 1, describing a disconnected and connected junction respectively.With the use of an adiabatic process starting from $\mathscr{H}_0$ at $t=-\infty$, it is easy to obtain the Keldysh NGF [8] for the central island orbital. We employ the Langreth-Wilkins convention[10] and construct the triplet of propagators G^R, G^A and the "less" particle correlation function $G^<$ of Kadanoff and Baym [9]. The Dyson equations for the propagators have the form

$$G^{R,A} = G_0^{R,A} + G_0^{R,A}\Sigma^{R,A}G^{R,A}, \text{ etc.} \tag{3}$$

while the less component satisfies the Dyson equation

$$G^< = G^R\Sigma^<G^A \tag{4}$$

All components of the self-energy have the form

$$\begin{aligned}\Sigma^X(t,t') &= \Sigma_L^X(t,t') + \Sigma_R^X(t,t')\\ \Sigma_Y^X(t,t') &= \alpha_Y(t)\sigma_Y^X(\tau)\alpha_Y(t')\\ \sigma_Y^{R,A}(\tau) &= \pm\mathrm{i}\int\tfrac{dE}{2\pi}\Delta_Y(E)\mathrm{e}^{-\mathrm{i}E\tau}\vartheta(\pm\tau)\\ \sigma_Y^<(\tau) &= \mathrm{i}\int\tfrac{dE}{2\pi}\Delta_Y(E)f_Y(E)\mathrm{e}^{-\mathrm{i}E\tau}\\ \tau &= t-t', \quad X=R,A,< \quad Y=L,R\end{aligned} \tag{5}$$

Each fully connected junction is thus associated with a spectral function Δ_Y which, together with the quasi-equilibrium Fermi function f_Y, specifies the Y self-energy triplet. Customarily, the spectral function is taken as flat (so-called wide band limit). In fact, this function is physically very similar to the adsorption function of Grimley or Newns [11] and may have a pronounced spectral structure which is transferred into the short time behavior of the self-energy[5].

In principle, any time-dependent process at the bridge is now solved. It is both productive and illuminating to make one more step, however, and view a transient starting at a finite time t_1 as embedded into the complete Keldysh "host" process[5]. This does not affect the propagators, but the Dyson equation for the restricted less function is now more involved [12,13]:

$$\begin{aligned}G_1^< &= G^R\Sigma_1^<G^A,\\ \Sigma_1^< &= \mathring{\Sigma}_1^< + \mathring{\Sigma}_1^{\bullet<} + \mathring{\Sigma}_1^{\circ<} + \mathring{\Sigma}_1^{\bullet\bullet<}\end{aligned} \tag{6}$$

Here, $\Sigma_1^<$ denotes the complete less self-energy of the transient, the symbolic multiplications involve integrations starting at t_1. The self-energy is modified so as to compensate for this restricted integration area. The four components of $\Sigma_1^<$ differ by the behavior at t_1, as indicated by circles. $\circ\cdots$ time variable fixed at t_1, $\bullet\cdots$ time variable continuous in (t_1,∞). Thus[5],

$$\begin{aligned}\mathring{\Sigma}_1^<(t,t') &= \mathrm{i}\rho_1(t_1)\delta(t-t_1-0)\delta(t'-t_1-0),\\ \mathring{\Sigma}_1^{\bullet<}(t,t') &= \mathring{\Lambda}_1^<(t_1,t')\delta(t-t_1-0),\\ \mathring{\Sigma}_1^{\circ<}(t,t') &= \mathring{\Lambda}_1^<(t,t_1)\delta(t'-t_1-0)\\ \mathring{\Sigma}_1^{\bullet\bullet<}(t,t') &= \Sigma^<(t,t') + \widehat{\Sigma}_1^<(t,t')\end{aligned} \tag{7}$$

The initial occupation $\rho(t_1)$ of the island orbital in the first line, and the host self-energy $\Sigma^<$ in the fourth line would describe the transient, if the initial state was an equilibrium state of the unperturbed Hamiltonian $\mathscr{H}_0$ (...a one-particle analogue of an uncorrelated initial condition). The additional quantities $\mathring{\Lambda}^<$, $\Lambda^{\circ<}$ and $\widehat{\Sigma}^<$ describe the initial correlation, or, put alternatively, the influence of past evolution on the commencing transient. We have developed a method of partitioning-in-time to construct these correction terms from the known NGF of the host process [14].

In the present case, the partitioning procedure can be extended into an iterative procedure. Let

$$-\infty \equiv t_0 \ll t_1 < t_2 \ \ldots\ < t_n < t_{n+1} \ \ldots \tag{8}$$

be the sequence of switching times with quiescent time intervals in between. We choose the t_n instants as the starting times of transient processes, the $n+1$th process embedded in the nth one. Then $G_{n+1}^<(t,t')$ for $t,t' > t_{n+1}$, and with lower integration limit t_{n+1}, becomes

$$\begin{aligned}G_{n+1}^< &= G^R\Sigma_{n+1}^<G^A,\\ \Sigma_{n+1}^< &= \mathring{\Sigma}_{n+1}^< + \mathring{\Sigma}_{n+1}^{\bullet<} + \mathring{\Sigma}_{n+1}^{\circ<} + \mathring{\Sigma}_{n+1}^{\bullet\bullet<}\end{aligned} \tag{9}$$

The self-energy is constructed from $G_n^<(t,t')$ known for $t,t' > t_n$. To avoid clumsy expressions, we illustrate the procedure by one example,

$$\begin{aligned}\mathring{\Lambda}_{n+1}^<(t_{n+1},u) &= \int_{t_n}^{t_{n+1}}\mathrm{i}\,\mathrm{d}\bar{t}\,\{G^R\Sigma_n^< + G_n^<\Sigma^A\}\\ G^R\Sigma_n^< &\mapsto G^R(t_{n+1},\bar{t})\Sigma_n^<(\bar{t},u), \text{ etc.}\end{aligned} \tag{10}$$

Using (5), (8) and prescribing the switching sequence, these expressions can be made much more explicit. Presently more important is to observe that actual integration limits in Eq. (10)

may be narrowed[14]: the function $\Sigma_n^<(t,t')$ is nearly zero for $|t-t'|>\tau_c$, a characteristic decay time of correlations. The integration in (10) and all related expressions will thus extend from $\max(t_n, t_{n+1}-\tau_c)$ to t_{n+1}. The resulting transient behavior will depend on the degree of overlap of the subsequent processes of correlation decay, as indicated by the relative position of $t_n+\tau_c$ and $t_{n+1}-\tau_c$.

3 Response to switching junctions on and off

For actual calculations, the metallic leads were taken as identical. The spectral function $\Delta_Y(E)$ was broad, but with a resonant structure (FWHM 0.3 eV). This, together with the step-like distribution function $f_Y(E)$ gave rise to τ_c e in the 10 fs range. The assumed sudden switching events, may thus simulate processes with femtosecond switching times. The leads had constant bias $\pm$ 0.5eV with respect to the island level, so that the charge flowed from the L lead to the R one on the whole. At the initial time $t_1=0$ of the transient, the level was empty and disconnected from both leads. For $t>t_1$, the currents through both junctions were calculated from the usual formulas [7], or their partitioning derivates.

3.1 Case A: Island suddenly connected to the leads

As the first **Case A**, we show the transient induced by switching both junctions on at $t_1=0$ (studied in the wide band limit in [4]). Fig. 1 shows initial oscillations of the currents J_L, J_R at both junctions, followed after elapsing about 20 fs by balanced steady currents. Correspondingly the rate of occupancy of the island level first oscillates and saturates to zero. A deeper insight is provided by Fig. 2 showing $G^<$ as a double time function. The crest line of the plot along the diagonal $t=t'$ is the occupation number $\rho(t)=\mathrm{i}G^<(t,t)$. It exhibits three stages: initial oscillatory transient, gradual saturation at controlled by the transport relaxation time and finally the steady state.

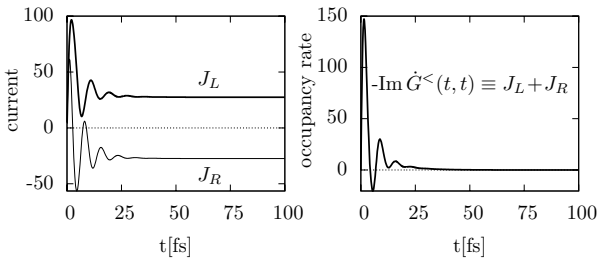

Fig. 1 Case A. Left panel: junction currents. Right panel: current balance at the island

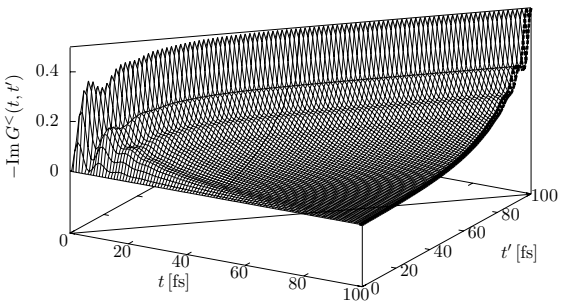

Fig. 2 Case A. Particle correlation function $G^<$. Darker part: steady state

3.2 Multiple switching

The next simplest **Case B** is specified by by the sequence of switching events: $t_1=0$ fs : L on, $t_2=40$ fs : R on, $t_3=70$ fs : R off, $t_4=80$ fs : R on. The L junction remains connected, while the R junction undergoes the switching on and off alternately.

In Figs. 3, 4, 5, the currents are shown for three subcases **B1, B2, B3**, whose label indicates the number of switch overs undergone by the R junction. As before, each switching induces first the transitory current oscillations, which die out gradually. The sign of the initial oscillation is such that the R junction "overreacts" at first and this is compensated later by the continued transient. It should be noted that the L junction invariably responds to the state of the R junction. Because the two junctions act differently having a mutually opposite bias, the L junction contributes to the compensation of the initial transient at the R junction. This is neatly demonstrated at the total occupancy rates. See Fig. 3 for **B1**. Turning the R junction off, Fig. 4 for **B2**, causes a transient reaction at L. The J_L current continues to flow by inertia at first and slows down in a wavy manner. Finally, in Fig. 5 for **B3**, we see how the next switch on coming early interrupts this process and the system returns to the final state of **B1**, but in an irregular manner caused by the overlap and mutual coupling of the two last transients.

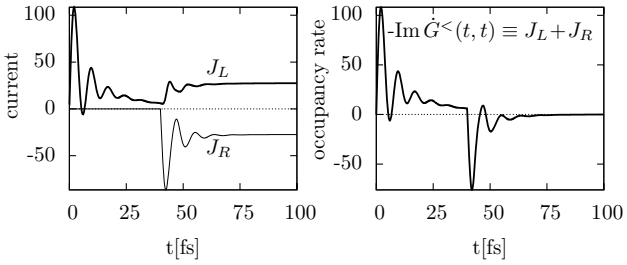

Fig. 3 Case B1 – see text. Left panel: junction currents. Right panel: current balance at the island

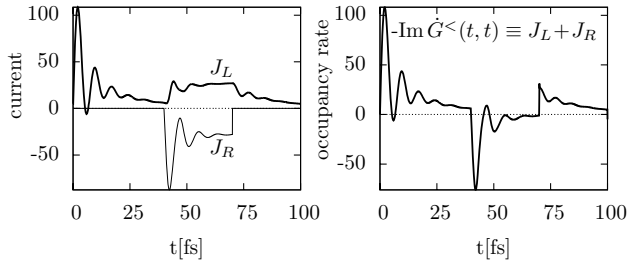

Fig. 4 Case B2 – see text. Left panel: junction currents. Right panel: current balance at the island

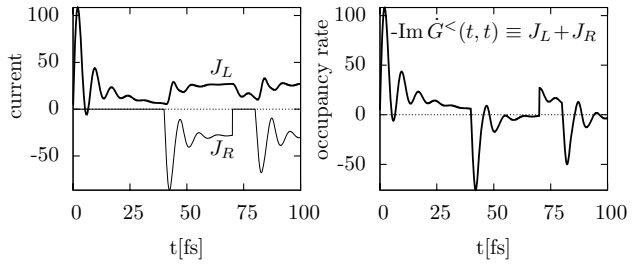

Fig. 5 Case B3 – see text. Left panel: junction currents. Right panel: current balance at the island

The richest **B3** process is further illuminated by Fig. 6 which displays the imaginary part of $G^<$ and can be compared with Fig. 2. The crest line represents the response of the island level occupation to the whole sequence of R switch-overs. The slopes of the whole plot (the symmetric one is hidden from the view) indicate the range of the coherent coupling of different areas. Clearly visible are the imprints of the $t = t_n$ and the $t' = t_n$ lines and the penetration of the coherent oscillations across these discontinuities.

It has been pointed out in Sec. 2 that the time partitioning permits to distinguish the correlated initial conditions from the uncorrelated ones by the presence of corrections to the less self-energy. This is demonstrated by Fig. 7 which shows one of the corrections, $\widehat{\Sigma}^<_n$. (see the last line of Eq. (7)). There is no correction at the leftmost corner $t = t' = t_1$, because there is no coherence between the past of the disconnected junction and the ensuing transient. By contrast, all later switch-overs give rise to a reconstruction of the initial state of the system and the correlation correction emerge. They die out soon, so that only the last pair, t_2 and t_3, appears to have the correlation correction overlapping and thus coupled.

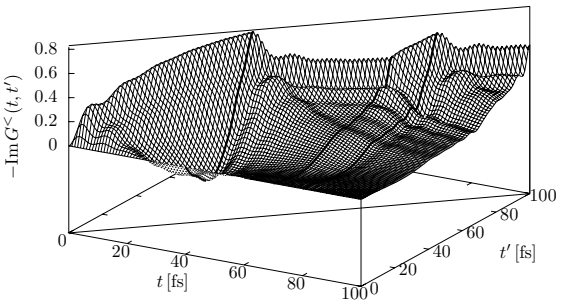

Fig. 6 Case B3. Particle correlation function $G^<$. See text

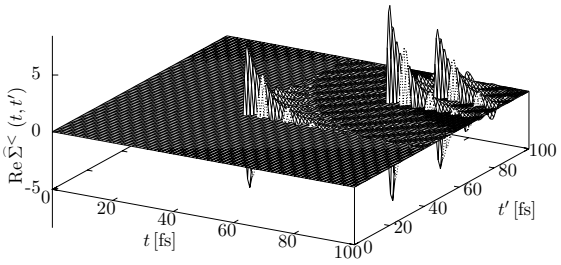

Fig. 7 Case B3. Correlation corrections $\widehat{\Sigma}^<$ to the lesser self-energy.

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