Transient electron currents through a molecular bridge in response to short switching sequences

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Bridge model (non-interacting electrons)

\[ Q_L = \sum_\ell |\ell\rangle\langle\ell| \quad \text{P} = |b\rangle\langle b| \quad Q_R = \sum_r |r\rangle\langle r| \]

Bound states decay into the continua of lead states
... genuine correlated behavior without interactions
Bridge model (non-interacting electrons)

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Bound states decay into the continua of lead states
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Analogy with interacting systems

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Bridge model (non-interacting electrons)

\[
\begin{align*}
Q_L &= \sum_{\ell} |\ell\rangle \langle \ell| \\
Q_R &= \sum_{r} |r\rangle \langle r| \\
P &= |b\rangle \langle b|
\end{align*}
\]

Bound states decay into the continua of lead states
... genuine correlated behavior without interactions

Analogy with interacting systems

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Q_L = \sum_\ell |\ell\rangle \langle \ell| \quad P = |b\rangle \langle b| \quad Q_R = \sum_r |r\rangle \langle r|
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<td>uncorrelated initial state</td>
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</tr>
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Bridge model (non-interacting electrons)

$\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}'(t)$

\[
\begin{align*}
\mathcal{H}' & = \mathcal{H}'_L + \mathcal{H}'_R \\
\text{coupling Hamiltonian}
\end{align*}
\]

Projectors

$\mathbf{1}_{\text{op}} = P + Q$, \hspace{1cm} $PQ = 0$

$P = \sum_b |b\rangle\langle b| \rightarrow |b\rangle\langle b|$

$Q = Q_L + Q_R \equiv \sum_\ell |\ell\rangle\langle \ell| + \sum_r |r\rangle\langle r|$

- discrete bound island states
- continuum of conducting lead states

Hamiltonian block structure

\[
\begin{align*}
\mathcal{H}_0 & = P \mathcal{H}_0 P + Q \mathcal{H}_0 Q \\
\mathcal{H}' & = P \mathcal{H}' Q + Q \mathcal{H}' P
\end{align*}
\]
NGF for a finite initial time

\[ G(1,1') = -i \langle T_c \psi(1) \psi^\dagger(1') \rangle_I \]

average over many-body initial state

arbitrary: non-equilibrium, with correlations...

\[ G \rightarrow \begin{pmatrix} G^R & G^< \\ 0 & G^A \end{pmatrix} \equiv G_{\text{LW}} \]

Reduction for a non-interacting system

\[ G^R = -i \mathcal{S}(t,t') \mathcal{G}(t-t') \]
\[ G^A = +i \mathcal{S}(t,t') \mathcal{G}(t'-t) \]
\[ G^<(t,t') = +i \mathcal{S}(t,t_1) \rho_1 \mathcal{S}(t_1,t') \]
\[ = +i G^R(t,t_1) \rho_1 G^A(t_1,t') \]

one-particle evolution operator

\[ i\hbar \frac{\partial}{\partial t} \mathcal{S}(t,t') = \mathcal{H}(t) \mathcal{S}(t,t') \quad \mathcal{S}(t,t) = 1 \]

one-particle \quad \uparrow \quad \text{Hamiltonian}

initial one-particle density matrix

\[ \rho_1(x,x') = \langle \psi(x,t_1) \psi^\dagger(x',t_1) \rangle_I \]
**NGF for the bound (island) states**

Bound states decay into continua of lead states
... genuine GF behavior without interactions

<table>
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<th>LEFT LEAD</th>
<th>ISLAND</th>
<th>RIGHT LEAD</th>
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<td>$Q_L = \sum_\ell</td>
<td>\ell\rangle\langle\ell</td>
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**Bound state Green’s functions**

$$G^R \rightarrow P G^R P = |b\rangle\langle b| G^R |b\rangle\langle b| = PG^R,$$

$$G^A \rightarrow P G^A P = |b\rangle\langle b| G^A |b\rangle\langle b| = PG^A,$$

$$G^< \rightarrow P G^< P = |b\rangle\langle b| G^< |b\rangle\langle b| = PG^<,$$

$$G^R \equiv \langle b | G^R | b \rangle$$

$$G^A \equiv \langle b | G^A | b \rangle$$

$$G^< \equiv \langle b | G^< | b \rangle$$
Dyson equations for propagators
by Löwdin partitioning
(folding down in Hilbert space)

Dyson equations for global propagators are unitary
\[ G^R = G^R_0 + G^R_0 \mathcal{H}' G^R, \quad G^R = G^R_0 + G^R \mathcal{H}' G^R_0 \]

Free propagators are block diagonal \( \Rightarrow \) partitioning expressions
\[ Q G^R |b\rangle = Q G^R_0 Q \mathcal{H}' |b\rangle G^R, \quad \langle b| G^R Q = G^R \langle b| \mathcal{H}' Q G^R_0 Q \]

Dyson equations for the island state propagators result
\[ G^R = G^R_0 + G^R_0 \sum^R G^R, \quad G^R = G^R_0 + G^R \sum^R G^R_0 \]
\[ \sum^R = \langle b| \mathcal{H}' Q G^R_0 Q \mathcal{H}' |b\rangle \]

- - - and the same for \( G^A \)
Dyson equations for propagators

by Löwdin partitioning

(folding down in Hilbert space)

\[ G^{<}(t, t') = \langle b | G^{<}(t, t') | b \rangle \]

\[ = +i \langle b | G^{R}(t, t_{1}) \varphi_{1} G^{A}(t_{1}, t') | b \rangle \]

\[ G^{<} = +i \langle b | G^{R} P + Q \varphi_{1} P + Q G^{A} | b \rangle \ldots \text{four terms} \]

\[ G^{<} = G^{R} \Xi^{<} G^{A} \]

\[ \Xi^{<} = \ldots i \rho(t_{1}) \delta_{tt_{1}} \delta_{t't_{1}} \equiv i \langle b | \varphi_{1} | b \rangle \delta_{tt_{1}} \delta_{t't_{1}} \]

\[ + \ldots \Lambda^{<}(t, t_{1}) \delta_{t't_{1}} \equiv i \langle b | H'Q G^{R}_{0}(t, t_{1})Q \varphi_{1} | b \rangle \delta_{t't_{1}} \]

\[ + \ldots \Lambda^{<}(t_{1}, t') \delta_{tt_{1}} \equiv i \langle b | \varphi_{1} Q G^{A}_{0}(t_{1}, t')Q H'| b \rangle \delta_{tt_{1}} \]

\[ + \ldots \Sigma^{<}(t, t') \equiv i \langle b | H'Q G^{R}_{0}(t, t_{1})Q \varphi_{1} Q G^{A}_{0}(t_{1}, t')Q H'| b \rangle \]

sensitive to initial conditions

by partitioning
Dyson equations for propagators by Löwdin partitioning (folding down in Hilbert space)

\[ G^<(t,t') = \langle b | G^<(t,t') | b \rangle = +i \langle b | G^R(t,t') \rho_1 G^A(t,t') | b \rangle \]

\[ G^<(t,t') = \int dt_1 \int dt_2 \ G^R(t,t) \mathcal{E}^<(t_1,t) G^A(t_2,t') \]

\[ \mathcal{E}^< = \ldots \mathcal{A}^<(t_1,t') \delta_{tt} \equiv i \langle b | \rho_1 Q G^A_0(t_1,t') Q \mathcal{H}' | b \rangle \delta_{tt} \]

\[ \ldots \mathcal{E}^< \equiv i \langle b | \mathcal{H}' Q G^R_0(t,t_1) Q \rho_1 Q G^A_0(t_1,t') Q \mathcal{H}' | b \rangle \]
Dyson equations for propagators by Löwdin partitioning (folding down in Hilbert space)

\[ G^< (t,t') = \langle b | G^< (t,t') | b \rangle = +i \langle b | G^R (t,t_I) \rho_I G^A (t_I,t') | b \rangle \]

\[ G^< = +i \langle b | G^R \ P + Q \ \rho_I \ P + Q \ G^A | b \rangle \ ... \ four \ terms \]

\[ E^< = E^< \ ... \ i \ \rho(t_I) \delta_{tt_I} \delta_{t't_I} \equiv i \langle b | \rho_I | b \rangle \delta_{tt_I} \delta_{t't_I} \]

\[ + \ E^< \ ... \ \Lambda^< (t,t_I) \delta_{t't_I} \equiv i \langle b | H' Q G^R_0 (t,t_I) Q \ \rho_I | b \rangle \delta_{t't_I} \]

\[ + \ E^< \ ... \ \Lambda^< (t_I,t') \delta_{tt_I} \equiv i \langle b | \rho_I \ Q G^A_0 (t_I,t') Q H' | b \rangle \delta_{tt_I} \]

\[ + \ E^< \ ... \ \tilde{\Sigma}^< (t,t') \equiv i \langle b | H' Q G^R_0 (t,t_I) Q \ \rho_I \ Q G^A_0 (t_I,t') Q H' | b \rangle \]
Dyson equations for propagators by Löwdin partitioning (folding down in Hilbert space)

\[ G^< = G^R \Xi^< G^A \]

\[ \Xi^< = \Xi^0 \quad \cdots \quad i \rho(t_1) \delta_{t_1} \delta_{t',t_1} \quad \equiv \quad i \langle b | \varphi_1 | b \rangle \delta_{tt_1} \delta_{t',t_1} \]
\[ + \Xi^< \quad \cdots \quad \Lambda^<(t,t_1) \delta_{t',t_1} \quad \equiv \quad i \langle b | \mathcal{H}' \mathcal{Q} G_0^R(t,t_1) \mathcal{Q} \varphi_1 | b \rangle \delta_{t',t_1} \]
\[ + \Xi^< \quad \cdots \quad \Lambda^<(t_1,t') \delta_{t_1} \quad \equiv \quad i \langle b | \varphi_1 \mathcal{Q} G_0^A(t_1,t') \mathcal{Q} \mathcal{H}' | b \rangle \delta_{tt_1} \]
\[ + \Xi^< \quad \cdots \quad \tilde{\Sigma}^<(t,t') \quad \equiv \quad i \langle b | \mathcal{H}' \mathcal{Q} G_0^R(t,t_1) \mathcal{Q} \varphi_1 \mathcal{Q} G_0^A(t_1,t') \mathcal{Q} \mathcal{H}' | b \rangle \]

Structure of self-energy \( \Xi^< \):
well known in the field theory

\( \Xi^0 \): uncorrelated initial condition
\( \Xi^< \): attributes of initial correlations
\( \cdots \): famous \( \Sigma^<_c \) and \( \Sigma^c \)
\( \Xi^< \): IC transient + regular part

Meaning of self-energy \( \Xi^< \):
for the non-interacting bridge

\( \Xi^0 \): projected one-electron distribution
\( \Xi^< \): depend on off-diag. "coherences"
\( \Xi^< \): virtual excursions into the leads
Uncorrelated initial condition
exploring the analogy

Uncorrelated initial condition is defined by

$$\langle b | \varphi_1 Q = 0, \quad Q \varphi_1 | b \rangle = 0$$

Initial distribution is block-diagonal \( \Rightarrow \) the island and lead states are only coupled dynamically, through \( \mathcal{H}' \), not by the initial condition.

As expected,

\[
\Lambda^<_o = \Lambda^<_i = 0 \quad \Rightarrow \\
\Xi^<_o = \Xi^<_i = 0 \quad \text{uncorrelated limit}
\]

The Dyson equation reduces to

\[
G^< = G^R i \rho_1 G^A + G^R \tilde{\Sigma}^< G^A
\]

initial (coherent) transient \quad "transport" backscatt. term

Dying out in time \quad Gliding with the running time
decay time \quad memory depth

\( \text{QP relaxation time} \)
Uncorrelated initial condition
exploring the analogy

Uncorrelated initial condition is defined by
\[ \langle b | \varphi_1 Q = 0, \quad Q \varphi_1 | b \rangle = 0 \]

Initial distribution is block-diagonal \( \Rightarrow \) the island and lead states are only coupled dynamically, through \( \mathcal{H}' \), not by the initial condition.

As expected,
\[ \Lambda_o^\Lambda = \Lambda^\Lambda = 0 \quad \Rightarrow \]
\[ \dot{\mathcal{E}}_o^\mathcal{E} = \mathcal{E}^\mathcal{E} = 0 \quad \text{uncorrelated limit} \]

The Dyson equation reduces to
\[ G^\Lambda = G^R i \rho_1 G^A + G^R \mathcal{S}^\Lambda \mathcal{S}^A \]

initial (coherent) transient "transport" backscatt. term
Dying out in time decay time
Gliding with the running time memory depth
QP relaxation time

\[ \text{initial decay of correlations remains} \]
Uncorrelated initial condition
when is the self-energy independent of initial time?

Self-energy

\[ \tilde{\Sigma}^< (t, t'; t_1) = i \langle b | \mathcal{H}' Q G_0^R (t, t_1) Q \rho_1 Q G_0^A (t_1, t') Q \mathcal{H}' | b \rangle \]

does not simplify for the uncorrelated initial condition \( \langle b | \rho_1 Q = 0, Q \rho_1 | b \rangle = 0 \)

Stronger diagonality condition

\[ \left[ \mathcal{H}_0 (t), \rho_1 \right] = 0, \quad t \geq t_1 \]

implies \( \left[ G_0^{R,A} (t, t'), \rho_1 \right] = 0, \quad t \geq t_1 \)

\[ \Sigma^< (t, t'; t_1) = i \langle b | \mathcal{H}' Q G_0^R (t, t_1) Q \rho_1 Q G_0^A (t_1, t') Q \mathcal{H}' | b \rangle \]

\[ = \langle b | \mathcal{H}' Q i (G_0^R (t, t') - G_0^A (t, t')) \rho_1 Q \mathcal{H}' | b \rangle \]

\[ t_1 \text{ drops out} \]

\[ = \langle b | \mathcal{H}' Q \mathcal{A}_0 (t, t') \rho_1 Q \mathcal{H}' | b \rangle \]

spectral density

• independent of initial time
• somewhat like the fluctuation-dissipation structure ... towards the KB Ansatz
Uncorrelated initial condition
exploring the analogy

Uncorrelated initial condition is defined by
\[ \langle b | \varphi_I Q = 0, \quad Q \varphi_I | b \rangle = 0 \]

Initial distribution is block-diagonal ⇒ the island and lead states are only coupled dynamically, through \( \mathcal{H}' \), not by the initial condition.

As expected,
\[ \Lambda_o^\leq = \Lambda^\leq = 0 \quad \Rightarrow \]
\[ \mathcal{E}^\leq = \mathcal{E}^\leq = 0 \quad \text{uncorrelated limit} \]

The Dyson equation reduces to
\[ G^\leq = G^R i \rho_1 G^A + G^R \Sigma^< G^A \]

initial (coherent) transient "transport" backscatt. term

Dying out in time decay time Gliding with the running time memory depth
QP relaxation time
Correlated initial condition
exploring the analogy

Correlated initial condition is defined by

$$\langle b|\varphi_1 Q \neq 0, \quad Q \varphi_1 |b \rangle \neq 0$$

Initial distribution is not block-diagonal ⇒ the island and lead states are coupled both dynamically, through $\mathcal{H}'$ and by the initial condition.

As expected,

$$A_0^\leq, \sigma A_0^\leq \neq 0 \quad \Rightarrow$$

$$\bar{E}_0^\leq, \sigma \bar{E}_0^\leq \neq 0 \quad \text{correlated limit}$$

The Dyson equation is complete

$$G^\leq = G^R i \rho_1 G^A + G^R A_0^\leq G^A + G^R \sigma A^\leq G^A + G^R \bar{\Sigma}^\leq G^A$$

initial (coherent) transient
Dying out in time
decay time

"transport" backscatt. term
Gliding with the running time
memory depth

QP relaxation time
Various approaches to correlated initial conditions

Two complementary techniques dealing with correlated initial conditions in current use are compared:

- those using characteristics of the initial state at $t_1$  Here ... the direct method
- those using the NGF along an extended Schwinger-Keldysh loop  Here ... the time partitioning
Two approaches to the correlated initial conditions

**Correlated initial conditions** have more recently been attacked along two complementary lines:

1. **the synchronous techniques**: the correlated initial state represented by a chain of correlation functions at a single initial time instant and suitably terminated

   *Klimontovich → Kremp → … → Bonitz & Semkat …*

2. **the diachronous techniques**: the finite-time Keldysh loop is extended, commonly by an imaginary stretch, the NGF determined along the extended contour starting at an uncorrelated state; either this is the result, or the finite-loop NGF is deduced by a contraction

   *Fujita → Hall → Danielewicz → … → Wagner → Morozov & Röpke …*
Diachronous vs. synchronous for our bridge model

1. the synchronous approach: represented by the direct solution by Hilbert space partitioning. In this special case, yields an exact explicit result expressing the self-energy in terms of the full initial density matrix. For example,

\[ \Lambda^<(t,t_1) = i \langle b | H' Q G^R_0(t,t_1) Q \rho_1 | b \rangle \]

2. the diachronous techniques: the finite-time Keldysh loop is extended along the real time axis now to incorporate the so-called switch-on states (next slide) starting at an uncorrelated state in the distant past. Only NGF enters along the extended contour and the finite-loop NGF is deduced by a subsequent time-partitioning.
Diachronous vs. synchronous for our bridge model

1. **The synchronous approach**: represented by the direct solution by Hilbert space partitioning. In this special case, yields an exact explicit result expressing the self-energy in terms of the full initial density matrix. For example,

\[ \Lambda^<_{\omega}(t,t_1) = i \langle b | \mathcal{H}' Q G^R_0(t,t_1) Q \varphi_1 | b \rangle \]

2. **The diachronous techniques**: the finite-time Keldysh loop is extended along the real time axis now to incorporate the so-called switch-on states (next slide) starting at an uncorrelated state in the distant past. Only NGF enters along the extended contour and the finite-loop NGF is deduced by a subsequent time-partitioning.
Initial states created by switch-on processes

A “physical” initial state is prepared at $t=t_1$ by a switch-on process antecedent to our transient. The initial conditions at this instant are fully captured by the NGF for the joint process \{preparation & transient\}. The transient NGF is extracted by a projection on times future with respect to $t_1$ (time partitioning).
Extension of the Keldysh loop
Extension of the Keldysh loop: past and future
Original Keldysh idea of switching on the interaction

Switch-on transient process with Keldysh initial condition

Uncorrelated initial state $t_{-\infty} \rightarrow -\infty$

Equilibrium

Correlated equil. state $t_I$ $t_U$

Pulse envelope

Transient process

Observation period

Time $t$
Arbitrary preparatory process in the past

Switch-on transient process with Keldysh initial condition

Switch-on process with Keldysh init. cond. and preparation stage
Dyson equation for particle correlation function

\[ G^{<}_{-\infty}(t, t') = \int_{-\infty}^{t} du \int_{-\infty}^{t'} dv G^{R}(u, v) \Sigma^{<} G^{A}(u, v) \]

\[ G^{<}(t, t') = \int_{t_1}^{t} du \int_{t_1}^{t'} dv G^{R}(u, v) \Xi^{<} G^{A}(u, v) \]
Dyson equation for particle correlation function

\[ G^<_{-\infty}(t, t') = \int_{-\infty}^t du \int_{-\infty}^{t'} dv G^R \Sigma^<G^A \]

\[ G^<_{-\infty} = G^R \sum^<G^A \]

\[ G^< (t, t') = \int_{t_1}^t du \int_{t_1}^{t'} dv G^R \Xi^<G^A \]

Different integration ranges !!!
Dyson equation for particle correlation function

**HOST PROCESS**

\[ G^{<}_\infty (t, t') = G^{R} \int_{-\infty}^{t} du \int_{-\infty}^{t'} dv G^{R} \Sigma^{<} G^{A} \]

**TRANSIENT**

\[ G^{<} (t, t') = G^{R} \Xi^{<} G^{A} \]

\[ \Xi^{<} = \Xi_{0}^{<} + \Lambda_{0}^{<}(t, t_{1}) \delta(t' - t_{1}) + \Lambda_{\ast}^{<}(t_{1}, t'_{1}) \delta(t - t_{1}) + \Xi_{\ast}^{<} \]

**NEW INTERPRETATION OF \( \Xi^{<} \):** the purple terms add to the host \( \Sigma^{<} \) in the right hand integral to compensate for the reduced integration range.

**TASK:** express the future-future block of \( \Xi^{<} \) in terms of the past-past and past-future blocks of the host GF and self-energies

**PARTITIONING-IN-TIME METHOD**
Time partitioning for self-energy

**EXAMPLE**

\[
\Lambda^\leq(v,t_1) = (-i) \int_{-\infty}^{t_1} d\bar{t} \left\{ \Sigma^\leq(v,\bar{t})G^A(\bar{t},t_1) + \Sigma^R(v,\bar{t})G^\leq_{-\infty}(\bar{t},t_1) \right\}
\]

- Diachronous form: only GF and self-energies of the island state occur, but for arbitrary times
- the GF (propagation) entirely in the past, the self-energies bridging over the initial time -- linking the past with the future
Time partitioning for self-energy \( \text{vs. the direct method} \)

**EXAMPLE**

\[
\Lambda_<(v,t_1) = (-i) \int_{-\infty}^{t_1} d\bar{t} \left\{ \Sigma^<(v,\bar{t})G^A(\bar{t},t_1) + \Sigma^R(v,\bar{t})G^<_R(\bar{t},t_1) \right\}
\]

- **Diachronous form:** only GF and self-energies of the island state occur, but for arbitrary times

- the GF (propagation) entirely in the past, the self-energies bridging over the initial time -- linking the past with the future

➤ Compare with synchronous approach: the direct solution by Hilbert space partitioning is

\[
\Lambda_<(t,t_1) = i\langle b|\mathcal{H}'QG^R_0(t,t_1)Q\varphi_1|b\rangle,
\]

- the two solutions can be derived one from the other -- verification of their equivalence, visualization of complementarity of both views.

**Basic idea:** employ the evolution in the past from the Keldysh initial cond.

\[
\Lambda_<(t,t_1) = i\langle b|\mathcal{H}'QG^R_0(t,t_1)Q\varphi_1|b\rangle,
\]

\[
G^R(t_1,t_{-\infty})\mathcal{P}_{-\infty} G^A(t_{-\infty},t_1)
\]

\(\mathcal{P}_{-\infty}\) block diagonal ("uncorrelated")

The rest is an algebra employing the Dyson equations for \(G^R, G^A\).
Time partitioning and decay of correlations

EXAMPLE

\[ \Lambda^{<}(v,t) = (-i) \int_{-\infty}^{t_1} d\bar{t} \left\{ \Sigma^{<}(v,\bar{t})G^{A}(\bar{t},t_1) + \Sigma^{R}(v,\bar{t})G^{<}_{-\infty}(\bar{t},t_1) \right\} \]

- Diachronous form: only GF and self-energies of the island state occur, but for arbitrary times
- the GF (propagation) entirely in the past, the self-energies bridging over the initial time -- linking the past with the future

Typical hierarchy of times

\[
\tau^* \ll \tau \ll \tau_H
\]

time depth of the self-energy \( \Sigma^{R,A,<}(t, t') \approx 0 \) for \( |t - t'| > \tau^* \)

thus

\[
t_1 - \tau^* < \bar{t} < t < v < t_1 + \tau^* \quad (Bogolyubov\ principle)
\]
Molecular bridge with time-dependent coupling

One source of transient behavior in the bridge structure are fast changes in the coupling strengths of both junctions. We contrast different sudden transitions between a coupled and an uncoupled state of a junction, some reducing to the uncorrelated initial state, other manifesting the correlated behavior.
Changes in the coupling strength of the junctions are interesting
• can be very fast – extreme of possibility of ensuing transients
• exotic – represent a time dependent “interaction” strength in our analogy
• partitioning in time is technically well suited

Bridge Hamiltonian further specialized

\[ \mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}'(t) \]
\[ \mathcal{H}'(t) = \alpha_R(t) \mathcal{V}_R + \alpha_L(t) \mathcal{V}_L \]

All time dependence in the \( \alpha \) amplitudes

Processes start as switch-on at \( t_{-\infty} \rightarrow -\infty \),
the couplings switched on adiabatically in the past (preparation),
then vary arbitrarily starting from \( t_1 \) (transient – observation)
Expressions for self-energy of the switching processes

All components of the self-energy have only regular parts (... $t_{-\infty} \rightarrow -\infty$) identical with usual JWM expressions:

$$
\Sigma^X(t,t') = \langle b | \mathcal{H}'(t) Q G_0^X Q \mathcal{H}'(t') | b \rangle
$$

$$
= \alpha_L(t) \sigma_L^X(t,t') \alpha_L(t') + \alpha_R(t) \sigma_R^X(t,t') \alpha_R(t')
$$

$$
\sigma_Y^X(t,t') = \langle b | \mathcal{V}_Y Q G_0^X(t,t') Q \mathcal{V}_Y | b \rangle,
$$

$X = R, A, <$

$Y = L, R$

$\mathcal{H}'(t) = \alpha_R(t) \mathcal{V}_R + \alpha_L(t) \mathcal{V}_L$
Expressions for self-energy of the switching processes

All components of the self-energy have only regular parts \((... t_{-\infty} \to -\infty)\) identical with usual JWM expressions:

\[
\Sigma^X(t,t') = \langle b | \mathcal{H}'(t) Q G_0^X Q \mathcal{H}'(t') | b \rangle = \alpha_L(t) \sigma^X_L(t,t') \alpha_L(t') + \alpha_R(t) \sigma^X_R(t,t') \alpha_R(t')
\]

\[
\sigma^X_Y(t,t') = \langle b | \mathcal{V}_Y Q G_0^X(t,t') Q \mathcal{V}_Y | b \rangle, \quad X = R, A, < \quad Y = L, R
\]

These “host process” steady single junction self-energies will serve as building blocks for generating the transient self-energies by partitioning-in-time.
Expressions for self-energy of the switching processes

All components of the self-energy have only regular parts ( \( \ldots t_{-\infty} \rightarrow -\infty \) ) identical with usual JWM expressions:

\[
\Sigma^X (t,t') = \langle b | \mathcal{H}'(t) Q G_0^X Q \mathcal{H}'(t') | b \rangle = \alpha_L(t) \sigma^X_L (t,t') \alpha_L(t') + \alpha_R(t) \sigma^X_R (t,t') \alpha_R(t')
\]

\[
\sigma^X_Y (t,t') = \langle b | \mathcal{V}_Y Q G_0^X (t,t') Q \mathcal{V}_Y | b \rangle,
\]

These “host process” steady single junction self-energies will serve as building blocks for generating the transient self-energies by partitioning-in-time. \( \rightarrow \) Transformation to Fourier integrals of coupling spectral densities

\[
\sigma^{R,A}_Y (t,t') = \pm i \langle b | \mathcal{V}_Y Q \mathcal{A}_0 (t,t') Q \mathcal{A}_0 Q \mathcal{V}_Y | b \rangle \int (\pm (t-t'))
\]

\[
\sigma^<_Y (t,t') = i \langle b | \mathcal{V}_Y Q \mathcal{A}_0 (t,t') \mathcal{V}_Y | b \rangle
\]

\[
\sigma^{R,A}_Y (t,t') = \pm i \int \frac{dE}{2\pi} \Delta_Y (E) e^{-iE(t-t')} \int (\pm (t-t'))
\]

\[
\sigma^<_Y (t,t') = i \int \frac{dE}{2\pi} \Delta_Y (E) f_Y (E) e^{-iE(t-t')}
\]

\[
\Delta_Y (E) = \frac{\Delta_Y (E)}{2\pi} \langle b | \mathcal{V}_Y \delta (E - \mathcal{H}_{0Y}) \mathcal{V}_Y | b \rangle
\]
Model electronic structure used in calculations

\[ Q_L = \sum_\ell |\ell\rangle\langle\ell| \quad P = |b\rangle\langle b| \quad Q_R = \sum_r |r\rangle\langle r| \]

The resonant coupling spectral density floating at the Fermi level

... Newns model of transition metal substrate
Model electronic structure used in calculations

\[
Q_L = \sum_{\ell} \langle \ell | \ell \rangle \\
Q_R = \sum_{r} \langle r | r \rangle \\
P = |b\rangle \langle b|
\]

\(\Delta_L(E)\)

\(\Delta_R(E)\)

\(f_L(E)\)

\(f_R(E)\)

The resonant coupling spectral density floating at the Fermi level

… Newns model of transition metal substrate

Compare with the WBL model sp. density (wide band limit) for the left electrode
Two leads connected simultaneously
Uncorrelated initial condition case

\[ -(\dot{N}_- + \dot{N}_+) = \dot{G}^<(t, t) \]

\[
\dot{N}_+ = -\frac{i}{\hbar} \text{Im} \left\{ \int_{t_0}^{t} \Sigma_-^R G_+^< + \int_{t_0}^{t} \Sigma_-^A G_+^A \right\} \\
\dot{N}_- = -\frac{i}{\hbar} \text{Im} \left\{ \int_{t_0}^{t} \Sigma_+^R G_-^< + \int_{t_0}^{t} \Sigma_+^A G_-^A \right\}
\]
Two leads connected simultaneously
Uncorrelated initial condition case

\[ \mathcal{G}_{++}^{-}(t, t') = \mathcal{G}_{++}^{R} \rho_0 \mathcal{G}_{++}^{A} + \mathcal{G}_{++}^{R} \Sigma_{++}^{<} \mathcal{G}_{++}^{A} \]
Two leads connected simultaneously
Uncorrelated initial condition case

decoupled $t_0$ R+L coupled running time $t$

("Keldysh initial time")

steady state transport: Meir-Wingreen formula

$$J_{i=\pm,i=} = \frac{ie}{\hbar} \int \frac{d\epsilon}{2\pi} \Gamma_i(\epsilon - e\varphi_i) \left( G^<(\epsilon) + \int_{F_D,i} (\epsilon - e\varphi_i)[G^R(\epsilon) - G^A(\epsilon)] \right)$$

$$\Gamma_i = i(\Sigma_i^R - \Sigma_i^A) \quad G^{R,A,<} \equiv G^{R,A,<}$$
Two leads connected one by one

Uncorrelated IC

Correlated IC

\[ -(\dot{N}_\downarrow + \dot{N}_\uparrow) = \dot{G}^<(t,t) \]

\[ \dot{N}_\uparrow = -\frac{i}{\hbar} \text{Im}\{ \int_{t_0}^{t} \Sigma^R_{\downarrow} \dot{G}^< + \int_{t_0}^{t} \Sigma^{<\downarrow} G^A \} \]

\[ \dot{N}_\downarrow = -\frac{i}{\hbar} \text{Im}\{ \int_{t_0}^{t} \Sigma^R_{\uparrow} \dot{G}^< + \int_{t_0}^{t} \Sigma^{<\uparrow} G^A \} \]
Two leads connected one by one

Uncorrelated IC

Correlated IC

decoupled

$L$ coupled

$R+L$ coupled

($\text{"Keldysh initial time"}$)

(initial time)

running time $t$

\[
g^<(t, t') = g^R \rho_0 g^A + g^R \sum< g^A
\]
Two leads connected one by one

Uncorrelated IC

Correlated IC

decoupled

$t_0$

L coupled

$t_I$

R+L coupled

running time $t$

("Keldysh initial time")

initial time

\[
g^<(t, t') = \mathcal{G}^R \rho_0 \mathcal{G}^A + \mathcal{G}^R \sum^< \mathcal{G}^A
\]
Two leads connected one by one

Uncorrelated IC

Correlated IC

decoupled

$t_0$

L coupled

$t_l$

R+L coupled

running time $t$

("Keldysh initial time")

host process...L coupled

embedded process...L+R coupled
Two leads connected one by one

Uncorrelated IC  Correlated IC

decoupled  \( t_0 \)  L coupled  \( t_I \)
(“Keldysh initial time”)  \( t_I \)  R+L coupled  running time \( t \)

Two leads connected one by one

Uncorrelated IC  Correlated IC

decoupled  \( t_0 \)  L coupled  \( t_I \)
(“Keldysh initial time”)  \( t_I \)  R+L coupled  running time \( t \)

\( t, t' > t_I \)

\[
\mathcal{G}^<(t, t') = \\
\mathcal{G}^R \rho_I \mathcal{G}^A + \mathcal{G}^R \Lambda^< \mathcal{G}^A + \mathcal{G}^R \Lambda^< \mathcal{G}^A + \mathcal{G}^R (\Sigma^< + \tilde{\Sigma}^<) \mathcal{G}^A
\]
Two leads connected one by one

Uncorrelated IC

Correlated IC

decoupled

$L$ coupled

$R+L$ coupled

initial time

running time $t$

$G^{<}(t, t') = G^{R} \rho_{I} G^{A} + G^{R} \Lambda_{o}^{<} G^{A} + G^{R} \Lambda^{<} G^{A} + G^{R} (\Sigma_{+}^{<} + \tilde{\Sigma}^{<}) G^{A}$
Two leads connected one by one

Uncorrelated IC  Correlated IC

decoupled  \( t_0 \)  L coupled  \( t_I \)  R+L coupled  running time \( t \)

(“Keldysh initial time”)

\[
\overline{A}^< = \int_{t_0}^{t_I} (\mathcal{G}_1^R \Sigma_1^< - \mathcal{G}_1^< \Sigma_1^A)
\]

\[
\Lambda_o^< = \int_{t_0}^{t_I} (\Sigma_1^< \mathcal{G}_1^A + \Sigma_1^R \mathcal{G}_1^<)
\]

\[
\tilde{\Sigma}^< = \frac{i}{\hbar} \int_{t_0}^{t_I} \int_{t_0}^{t_I} (-\Sigma_1^R \Sigma_1^< \mathcal{G}_1^R + \Sigma_1^R \mathcal{G}_1^< \Sigma_1^A + \Sigma_1^< \mathcal{G}_1^A \Sigma_1^A)
\]

\( t, t' > t_I \)

\[
\mathcal{G}^< (t, t') =
\]

\[
\mathcal{G}^R \rho_I \mathcal{G}^A + \mathcal{G}^R \overline{A}^< \mathcal{G}^A + \mathcal{G}^R \Lambda_o^< \mathcal{G}^A + \mathcal{G}^R (\Sigma^<_1 + \tilde{\Sigma}^<_1) \mathcal{G}^A
\]
Two leads connected one by one

**Uncorrelated IC**

- decoupled
- \( t_0 \) ("Keldysh initial time")

**Correlated IC**

- \( t_I \) initial time
- \( R+L \) coupled

\[ t, t' > t_I \]

\[
\mathcal{G}_{\text{tot}}^{<}(t, t') =
\mathcal{G}^{R} \rho_{I} \mathcal{G}^{A} + \mathcal{G}^{R} \mathcal{A}^{<} \mathcal{G}^{A} + \mathcal{G}^{R} \Lambda_{o}^{<} \mathcal{G}^{A} + \mathcal{G}^{R} \left( \Sigma^{<} + \Sigma^{<} \right) \mathcal{G}^{A}
\]
Two leads connected one by one

Uncorrelated IC

Correlated IC

decoupled

$L$ coupled

R+L coupled

running time $t$

("Keldysh initial time")

$t_0$

$t_I$

initial time

$t, t' > t_I$

\[
\mathcal{G}^{<}(t, t') = \mathcal{G}_{\text{UNCOR}}^{R} \rho_{I} \mathcal{G}^{A} + \mathcal{G}^{R} \mathcal{A}^{<} \mathcal{G}^{A} + \mathcal{G}^{R} \mathcal{A}_{O}^{<} \mathcal{G}^{A} + \mathcal{G}^{R} (\sum^{<} + \tilde{\sum}^{<}) \mathcal{G}^{A}
\]
Two leads connected one by one; finally R-off

- Uncorrelated IC
- Correlated IC
- Correlated IC

Decoupled $t_0$ ("Keldysh initial time")
L coupled $t_I$
R+L coupled $t_x$

Initial time initial time

Running time $t$

Graphs showing current over time $t$ [fs]:
- $\hat{N}_{-}$
- $\hat{g}^{<}(t, t)$

Graphs showing current over time $t$ [fs]:
- $\hat{N}_{-} + N_{-}$
Two leads connected one by one; finally R-off

- **Uncorrelated IC**
  - decoupled
  - $t_0$ ("Keldysh initial time")

- **Correlated IC**
  - L coupled
  - $t_I$ initial time
  - R+L coupled
  - $t_x$ initial time

Running time $t$

Diagram showing time-dependent transport with $g^<(t, t')$.
Two leads connected one by one; finally R-off

Uncorrelated IC

decoupled $t_0$

("Keldysh initial time")

L coupled $t_l$

initial time

Correlated IC

R+L coupled $t_x$

initial time

running time $t$
Correlated initial condition for an embedded process by time partitioning

Single molecule bridge as a testing ground for using NGF outside of the steady current regime

A. Kalvová, V. Špička, B. Velický: Non-equilibrium Statistical Physics Today
Fast transients in mesoscopic systems

Time-dependent transport in interacting and noninteracting resonant-tunneling systems

Time-dependent transport in interacting and noninteracting resonant-tunneling systems