Time-dependent transport through a nanostructure via the generalized master equation, II

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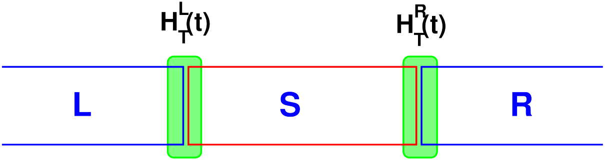
Coupling Hamiltonian

Before opening up the system to leads we need a coupling Hamiltonian



We want sensitivity to geometry

$$T^l_{aq} = \int_{\Omega^l_S \times \Omega_l} d\mathbf{r}\, d\mathbf{r}' \left(\psi^l_q(\mathbf{r}')\right)^* \psi^S_a(\mathbf{r}) g^l_{aq}(\mathbf{r}, \mathbf{r}')$$

Choice of domains and "overlap"

The integration domains for the leads are chosen to be

$$\begin{aligned}
\Omega_{\rm L} &= \left\{(x,y)\left|\left[-\frac{L_x}{2}-2a_w,-\frac{L_x}{2}\right]\times[-3a_w,+3a_w]\right.\right\},\\
\Omega_{\rm R} &= \left\{(x,y)\left|\left[+\frac{L_x}{2},+\frac{L_x}{2}+2a_w\right]\times[-3a_w,+3a_w]\right.\right\}
\end{aligned}$$

and for the system

$$\begin{aligned}
\Omega_{\rm S}^{\rm L} &= \left\{(x,y)\left|\left[-\frac{L_x}{2},-\frac{L_x}{2}+2a_w\right]\times[-3a_w,+3a_w]\right.\right\},\\
\Omega_{\rm S}^{\rm R} &= \left\{(x,y)\left|\left[+\frac{L_x}{2}-2a_w,+\frac{L_x}{2}\right]\times[-3a_w,+3a_w]\right.\right\}
\end{aligned}$$

The nonlocal overlap

$$g_{aq}^l(\mathbf{r},\mathbf{r}') = g_0^l \exp\left[-\delta_1^l(x-x')^2-\delta_2^l(y-y')^2\right]\exp\left(\frac{-|E_a-\epsilon^l(q)|}{\Delta_E^l}\right)$$

Many-electron coupling

We have T^l_{aq} coupling single-electron states (SESs), but we need $\mathcal{T}^l_{\mu\nu}(q)$ coupling many-electron states (MESs)

The Hamiltonian for the coupling is

$$H^l_{\mathrm{T}}(t) = \chi^l(t) \sum_{qa} \left\{ T^l_{qa} c^\dagger_{ql} d_a + (T^l_{qa})^* d^\dagger_a c_{ql} \right\}$$

- creation operator $c^\dagger_{ql}$ of electron in state $|q) = |n^l, q)$ in lead l
- creation operator $d^\dagger_a$ of electron in state $|a)$ in the system
- $\chi^l(t)$ switching function in time
- $\sum_q$ is summation over n^l and integration over q

We transform the Hamiltonian

$$\sum_{qa} T^l_{qa} c^\dagger_{ql} d_a = \sum_{\mu\nu qa} T^l_{qa} c^\dagger_{ql} |\mu\rangle\langle\mu| d_a |\nu\rangle\langle\nu|$$
$$= \sum_{\mu\nu q} |\mu\rangle c^\dagger_{ql} \left\{ \sum_a T^l_{qa} \langle\mu| d_a |\nu\rangle \right\} \langle\nu|$$
$$= \sum_{\mu\nu q} |\mu\rangle c^\dagger_{ql} \left(\mathcal{T}^l_{\mu\nu}(q) \right)^* \langle\nu| = \sum_q \left(\mathcal{T}^l(q) \right)^* c^\dagger_{ql}$$

where we define

$$\left(\mathcal{T}^l_{\mu\nu}(q) \right)^* = \sum_a T^l_{qa} \langle\mu| d_a |\nu\rangle$$
$$\left(\mathcal{T}^l(q) \right)^* = \sum_{\mu\nu} \left(\mathcal{T}^l_{\mu\nu}(q) \right)^* |\mu\rangle\langle\nu|$$

The coupling Hamitonian becomes

$$H_{\mathrm{T}}^{l}(t) = \chi^{l}(t) \sum_{ql} \left\{ \mathcal{T}^{l}(q) c_{ql} + c_{ql}^{\dagger} \left(\mathcal{T}^{l}(q) \right)^{*} \right\}$$

The Hamiltonian of the leads is

$$H^{l} = \sum_{q} \epsilon^{l}(q) c_{ql}^{\dagger} c_{ql}$$

In equilibrium, before coupling at $t = 0$ we have the density operator (probability operator)

$$\rho_{l} = \frac{e^{-\beta(H_{l} - \mu_{l} N_{l})}}{\mathrm{Tr}_{l}\{e^{-\beta(H_{l} - \mu_{l} N_{l})}\}}$$

$$c_{ql}(t) = e^{itH^{l}/\hbar} c_{ql} e^{-itH^{l}/\hbar} = c_{ql} e^{-it\epsilon(q)/\hbar}$$

and

$$\mathrm{Tr}_{l} \left\{ c_{ql}(t) c_{ks}^{\dagger}(t') \right\} = \delta(q-k) \delta_{l,s} e^{-i(t-t')\epsilon^{l}(q)/\hbar} \left\{ 1 - f(\epsilon^{l}(q)) \right\}$$

and

$$\mathrm{Tr}_l \left\{ c_{ql}^{\dagger}(t) c_{ks}(t') \right\} = \delta(q-k)\delta_{l,s} e^{+i(t-t')\epsilon^l(q)/\hbar} f(\epsilon^l(q))$$

$f(\epsilon^l(q))$ is the equilibrium distribution in lead l before $t=0$

We can transform the coupling tensor into the Coulomb interacting many-electron basis $\{|\mu)\}$

$$\tilde{\mathcal{T}}^l(q) = \mathcal{V}^{+}\mathcal{T}^l(q)\mathcal{V}, \quad \left(\tilde{\mathcal{T}}^l(q)\right)^* = \mathcal{V}^{+}\left(\mathcal{T}^l(q)\right)^*\mathcal{V}$$

Now we need to describe the evolution of the system after the coupling to the leads at $t=0$

Coupling to the leads - Derivation of the GME

Leads coupled to system at $t = 0$, nonequilibrium, electrons are into and through the system. We need to solve

$$\dot{W}(t) = -\frac{i}{\hbar}\left[H(t), W(t)\right] = -i\mathcal{L}(t)\, W(t)$$

with

$$H(t) = H_{\mathrm{S}} + H_{\mathrm{L}} + H_{\mathrm{R}} + H_{\mathrm{T}}(t)$$

Assume H_{S} is not time-dependent

Too large task, use projection formalism of Nakajima and Zwanzig
$\rightarrow$ time evolution of open system in presence of leads

Projection operators

System: H_S, reservoir: $H_\mathrm{res} = H_\mathrm{L} + H_\mathrm{R}$, coupling: $H_\mathrm{T}(t)$

Projection operators: $\mathcal{P} = \rho_\mathrm{res}(0)\mathrm{Tr}_\mathrm{res}$, $\mathcal{P} = 1 - \mathcal{Q}$

Reduced density operators

$$W(t) = \rho_{\mathrm{res+S}}(t), \quad \mathrm{Tr}_\mathrm{res}\rho_\mathrm{res}(0) = 1, \quad \mathcal{P}\,W(t) = \rho_\mathrm{res}(0)\rho_\mathrm{S}(t)$$

Mean value of an operator A

$$\langle A(t)\rangle = \mathrm{Tr}\{W(t)A\} = \mathrm{Tr}_\mathrm{S}\{\rho_\mathrm{S}(t)A\}$$

Operator properties

$$
\begin{aligned}
\mathcal{P}\mathcal{L}_{\mathrm{S}}\cdots &= \mathcal{L}_{\mathrm{S}}\mathcal{P}\cdots \\
\mathcal{P}\mathcal{L}_{\mathrm{res}}\cdots &\sim \mathcal{P}\left[H_{\mathrm{res}},\cdots\right] = 0 \\
\mathcal{Q}\mathcal{L}_{\mathrm{T}}\mathcal{P} &= 1-\mathcal{P}\mathcal{L}_{\mathrm{T}}\mathcal{P} = \mathcal{L}_{\mathrm{T}}\mathcal{P} - \underbrace{\mathcal{P}\mathcal{L}_{\mathrm{T}}}_{=0}\mathcal{P} = \mathcal{L}_{\mathrm{T}}\mathcal{P}
\end{aligned}
$$

$$
\begin{aligned}
\mathcal{P}\mathcal{Q} &= \mathcal{Q}\mathcal{P} = 0 \\
\mathcal{Q}\mathcal{L}_{\mathrm{S}} &= \mathcal{L}_{\mathrm{S}}\mathcal{Q} \\
\mathcal{Q}\mathcal{L}_{\mathrm{res}} &= \mathcal{L}_{\mathrm{res}}\mathcal{Q} = \mathcal{L}_{\mathrm{res}}
\end{aligned}
$$

H_{T} is linear in c_{ql} and $c_{ql}^{\dagger}$

$$
\begin{aligned}
\mathcal{P}\mathcal{L}\mathcal{P} &= \mathcal{L}_{\mathrm{S}}\mathcal{P} \\
\mathcal{P}\mathcal{L}\mathcal{Q} &= \mathcal{P}\mathcal{L}_{\mathrm{T}}\mathcal{Q} \\
\mathcal{Q}\mathcal{L}\mathcal{P} &= \mathcal{Q}\mathcal{L}_{\mathrm{T}}\mathcal{P} \\
\mathcal{Q}\mathcal{L}\mathcal{Q} &= \mathcal{L}_{\mathrm{S}}\mathcal{Q} + \mathcal{L}_{\mathrm{res}}\mathcal{Q} + \mathcal{Q}\mathcal{L}_{\mathrm{T}}\mathcal{Q}
\end{aligned}
$$

Equation of motion

We rewrite the equation of motion as

$$\mathcal{P}\,\dot{W}(t) = -i\mathcal{P}\mathcal{L}\mathcal{P}\,W - i\mathcal{P}\mathcal{L}\mathcal{Q}\,W$$
$$\mathcal{Q}\,\dot{W}(t) = -i\mathcal{Q}\mathcal{L}\mathcal{P}\,W - i\mathcal{Q}\mathcal{L}\mathcal{Q}\,W$$

and reduce to

$$\mathcal{P}\,\dot{W}(t) = -i\mathcal{L}_{\mathrm{S}}\mathcal{P}\,W - i\mathcal{P}\mathcal{L}_{\mathrm{T}}\mathcal{Q}\,W$$
$$\mathcal{Q}\,\dot{W}(t) = -i\mathcal{L}_{\mathrm{T}}\mathcal{P}\,W - i\left\{\mathcal{L}_{\mathrm{S}} + \mathcal{L}_{\mathrm{res}} + \mathcal{Q}\mathcal{L}_{\mathrm{T}}\right\}\mathcal{Q}\,W$$

Two coupled equations we solve under the condition of weak system leads coupling

Solution

The second equation has formal solution

$$\mathcal{Q}W(t) = -i\int_0^t dt'\ \mathrm{T}\exp\left\{-i\int_{t'}^t dt''\ \left[\mathcal{L}_{\mathrm{S}}(t'') + \mathcal{L}_{\mathrm{res}}(t'') + \mathcal{Q}\mathcal{L}_{\mathrm{T}}(t'')\right]\right\} \mathcal{L}_{\mathrm{T}}(t')\mathcal{P}W(t')$$

where we use $\mathcal{Q}W(0) = 0$, For weak coupling this becomes

$$\mathcal{Q}W(t) \approx -i\int_0^t dt'\ \exp\left\{-i\left[\mathcal{L}_{\mathrm{S}} + \mathcal{L}_{\mathrm{res}}\right](t-t')\right\}\mathcal{L}_{\mathrm{T}}(t')\mathcal{P}W(t')$$

as $\mathcal{L}_{\mathrm{S}}$ and $\mathcal{L}_{\mathrm{res}}$ do not depend on time

In the same approximation an explicit solution can be obtained to the inhomogeneous differential equation

$$\mathcal{Q}\,\dot{W}(t) \approx -i\mathcal{L}_{\rm T}\mathcal{P}\,W - i\left\{\mathcal{L}_0\right\}\mathcal{Q}\,W$$

as

$$\mathcal{Q}\,W(t) = -i\int_0^t dt'\;\, U_0(t-t')\mathcal{L}_{\rm T}(t')\mathcal{P}\,W(t')\,U_0^{+}(t-t')$$

where

$$\mathcal{L}_0 = \mathcal{L}_{\rm S} + \mathcal{L}_{\rm res}, \quad U_0(t) = e^{iH_0/\hbar}$$

By introducing $\rho_{\rm S}(t) = \mathcal{P}\,W(t)$ and combining with the first equation we get

$$\dot{\rho}_{\rm S}(t) = -i\mathcal{L}_{\rm S}\rho_{\rm S}(t) - \frac{1}{\hbar}\mathcal{P}\mathcal{L}_{\rm T}\int_0^t dt'\;\, U_0(t-t')\mathcal{L}_{\rm T}(t')\rho_{\rm S}(t')\,U_0^{+}(t-t')$$

or explicitly

$$\dot{\rho}_{\rm S}(t) = \frac{i}{\hbar}\left[H_{\rm S},\rho_{\rm S}(t)\right] - \frac{1}{\hbar^2}{\rm Tr}_{\rm res}\left\{\left[H_{\rm T}(t'),\int_0^t dt'\ \left[U_0(t-t')H_{\rm T}(t')U_0^+(t-t') \right.\right.\right.$$
$$\left.\left.\left. ,\ e^{-i(t-t')H_{\rm S}/\hbar}\rho_{\rm S}(t')e^{+i(t-t')H_{\rm S}/\hbar}\right]\right]\right\}$$

The generalized master equation (GME), an integro-differential equation with the kernel approximated to the second order in $H_{\rm T}$, not a Born approximation since the integral sturcture gives terms with $H_{\rm T}$ to any order

No Markov approximation $\rightarrow$ memory effects

No assumption about equilibrium in leads after coupling

Now we use our form of the coupling H_T to obtain

$$\begin{aligned}\dot{\rho}(t) &= -\frac{i}{\hbar}[H_\mathrm{S},\rho(t)] \\ &- \frac{1}{\hbar^2}\sum_{l=\mathrm{L,R}}\int dq\,\chi^l(t)([\mathcal{T}^l,\Omega_{ql}(t)]+h.c.)\end{aligned}$$

where two operators have been introduced to compactify the notation

$$\Omega_{ql}(t) = U_\mathrm{S}^\dagger(t)\int_{t_0}^{t} ds\,\chi^l(s)\Pi_{ql}(s)e^{i((s-t)/\hbar)\varepsilon^l(q)}\,U_\mathrm{S}(t),$$

$$\Pi_{ql}(s) = U_\mathrm{S}(s)\left(\mathcal{T}^{l\dagger}\rho(s)(1-f^l)-\rho(s)\mathcal{T}^{l\dagger}f^l\right)U_\mathrm{S}^\dagger(t),$$

with $U_\mathrm{S}(t)=e^{i(t/\hbar)H_\mathrm{S}}$, $f^l=f(\epsilon^l(q)-\mu_l)$

References

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