Time-dependent quantum transport through interacting systems with the Kadanoff-Baym equations

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Overview

- Aim and motivation

- Theoretical approach to quantum transport

    - The Kadanoff-Baym method
    - Correlated systems coupled to macroscopic leads
    - Results

- Conclusions

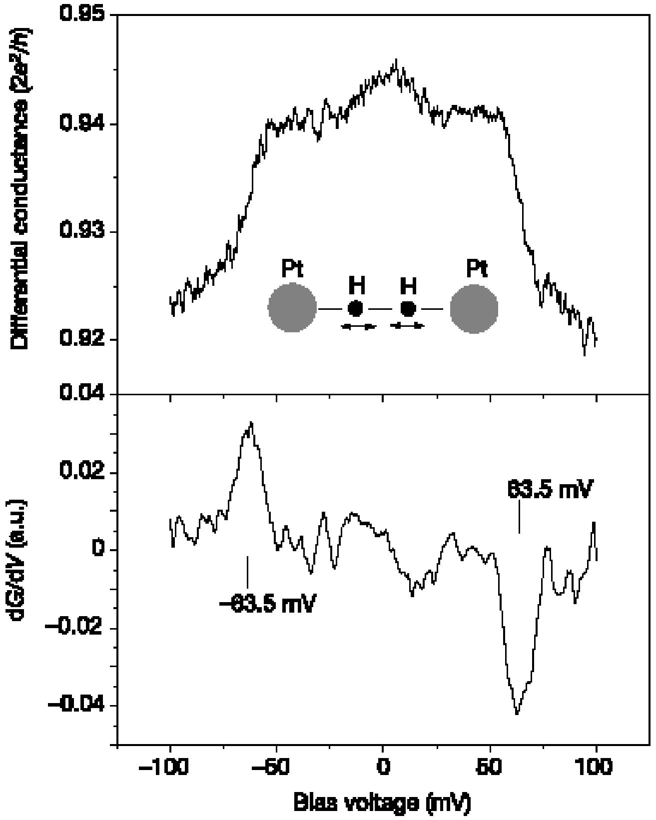
P.Myöhänen, A.Stan, G.Stefanucci, RvL

Phys.Rev. B80, 115107 (2009)
Europhys.Lett. 84, 67001 (2008)

Motivation

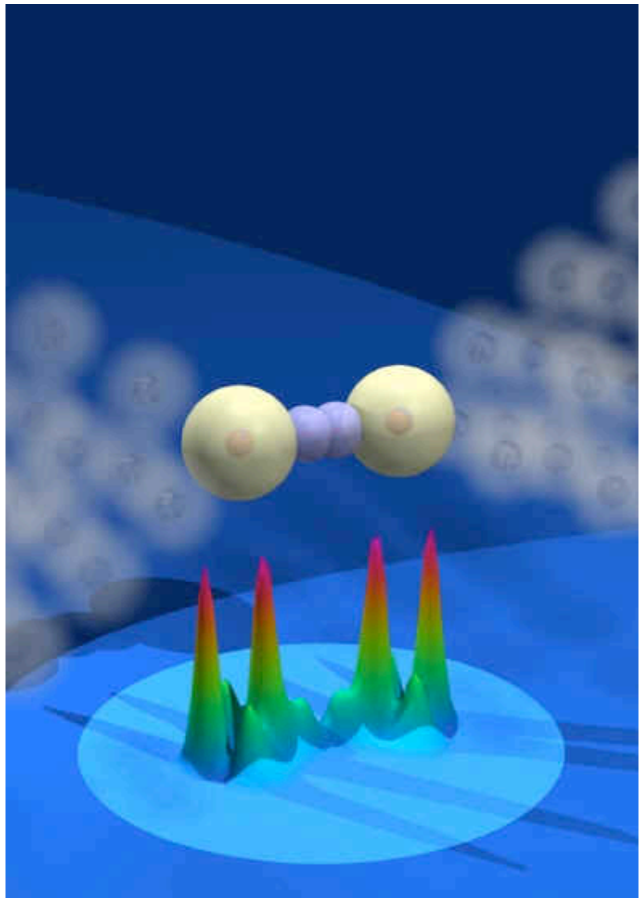
In nanoscience one aims to understand and control systems at the nanoscale.

Manipulation of nanosystems requires interactions with a time-dependent environment



Conduction through a single H_2 molecule

R.H.M.Smit et al.
Nature 419, 906 (2002)



The importance of dynamics

In future devices based on ‘molecular electronics’ we are not mainly interested in the steady states:
The operational speed needs to be designed and controlled

The main interest will be in fast switching of the devices!

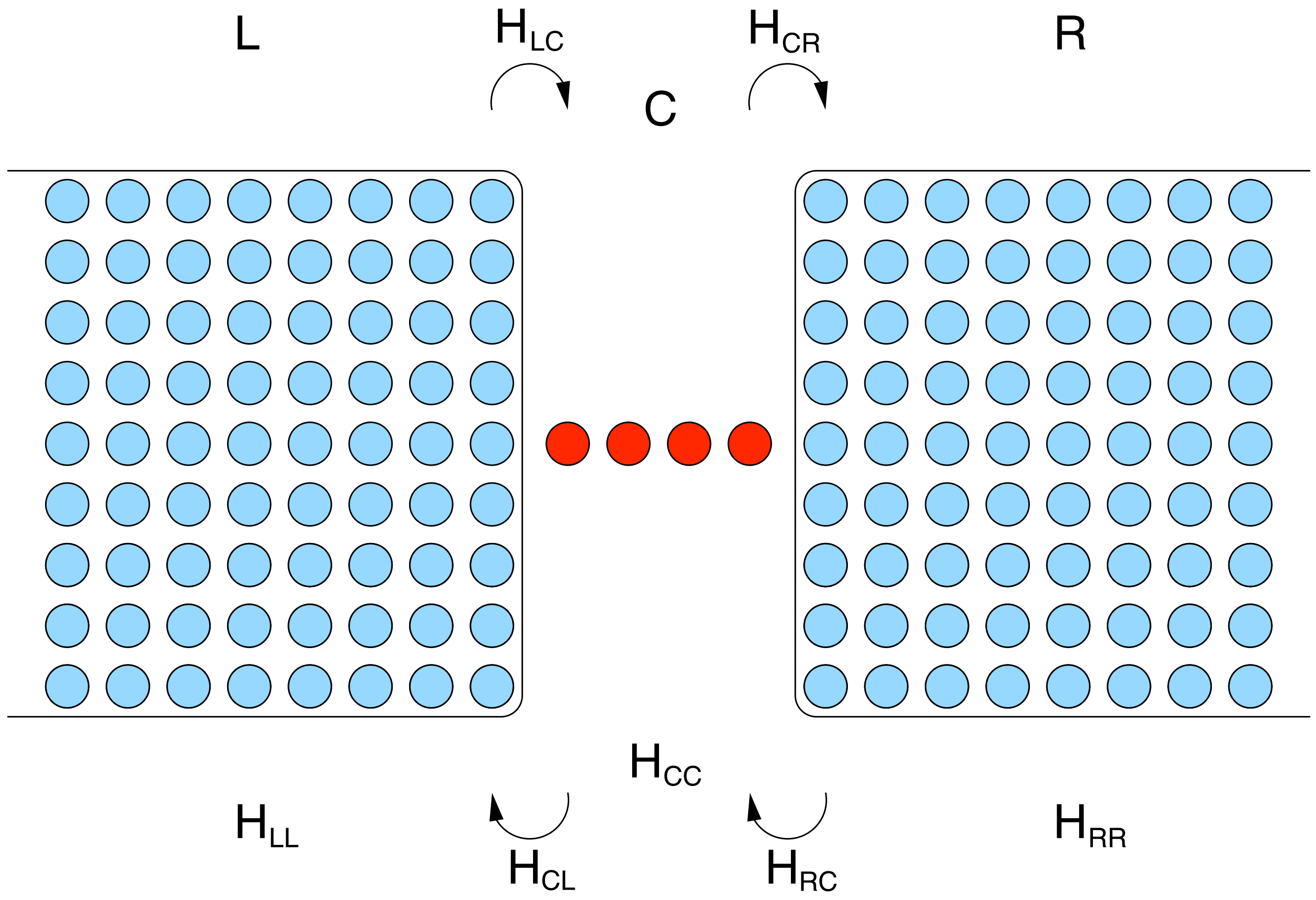
This involves:

- switching times (AC fields, lasers)
- study of transients
- peak currents rather than time-averaged ones (stability of the devices)

We need fundamental many-electron nonequilibrium quantum mechanics for open systems at short time-scales
(nice topic for the theorists with rich physics.....)

The time-dependent quantum transport problem

Consider a molecule (or quantum dot) attached to leads

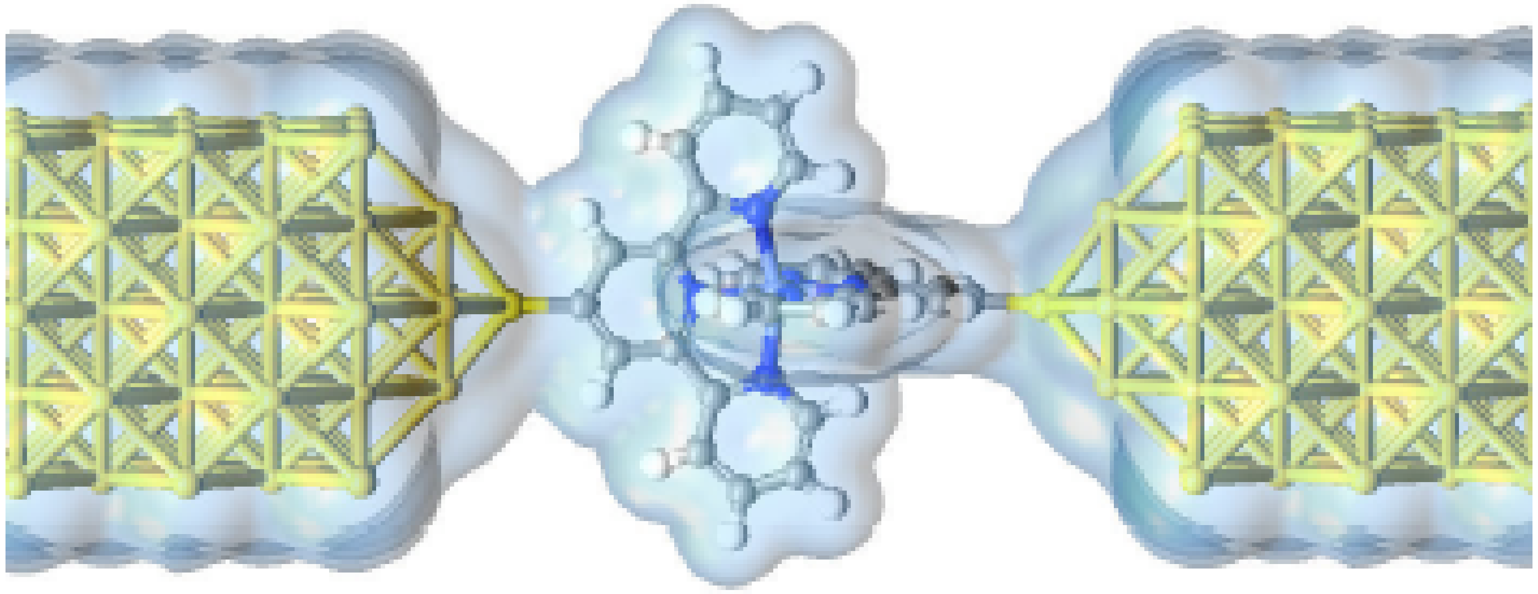


Problem:

Calculate the time evolution of observables of this system when a bias is applied.

Theoretical challenges

- We are dealing with an open quantum system
- We are dealing with a many-particle system
- We are dealing with a nonequilibrium system



Our approach

- We use Kadanoff-Baym approach to study the basic physics of time-dependent processes that play a role in correlated quantum transport
    - bistability, phonons, spin transport, superconducting leads, AC fields,.....etc.
- We use Kadanoff-Baym approach to benchmark and develop new correlation functionals in density functional theory beyond ALDA.

Time-propagation of the Kadanoff-Baym equations

(N.E. Dahlen, RvL,
Phys.Rev.Lett. 98, 153004 (2007)

P.Myöhänen, A.Stan, G.Stefanucci, RvL
Europhys.Lett. 84, 67001 (2008))

P.Myöhänen, A.Stan, G.Stefanucci, RvL
Phys.Rev. B80, 115107 (2009),

This method can deal with:

- inhomogeneous open systems
- time-dependent external fields
  (both in leads and device regions)
- electronic interactions

The method is based on the propagation of the equations of motion for the nonequilibrium Green function, also known as the Kadanoff-Baym equations. (Kadanoff/Baym 1964, Keldysh 1965)

The main ingredient of the method is the nonequilibrium Green function

$$G(\mathbf{x}_1 t_1, \mathbf{x}_2 t_2)$$

Time evolution of a many-body system

The time-dependent Hamiltonian

$$\hat{H}(t) = \hat{h}(t) + \hat{W}$$

Kinetic energy + external potential

Two-particle interactions

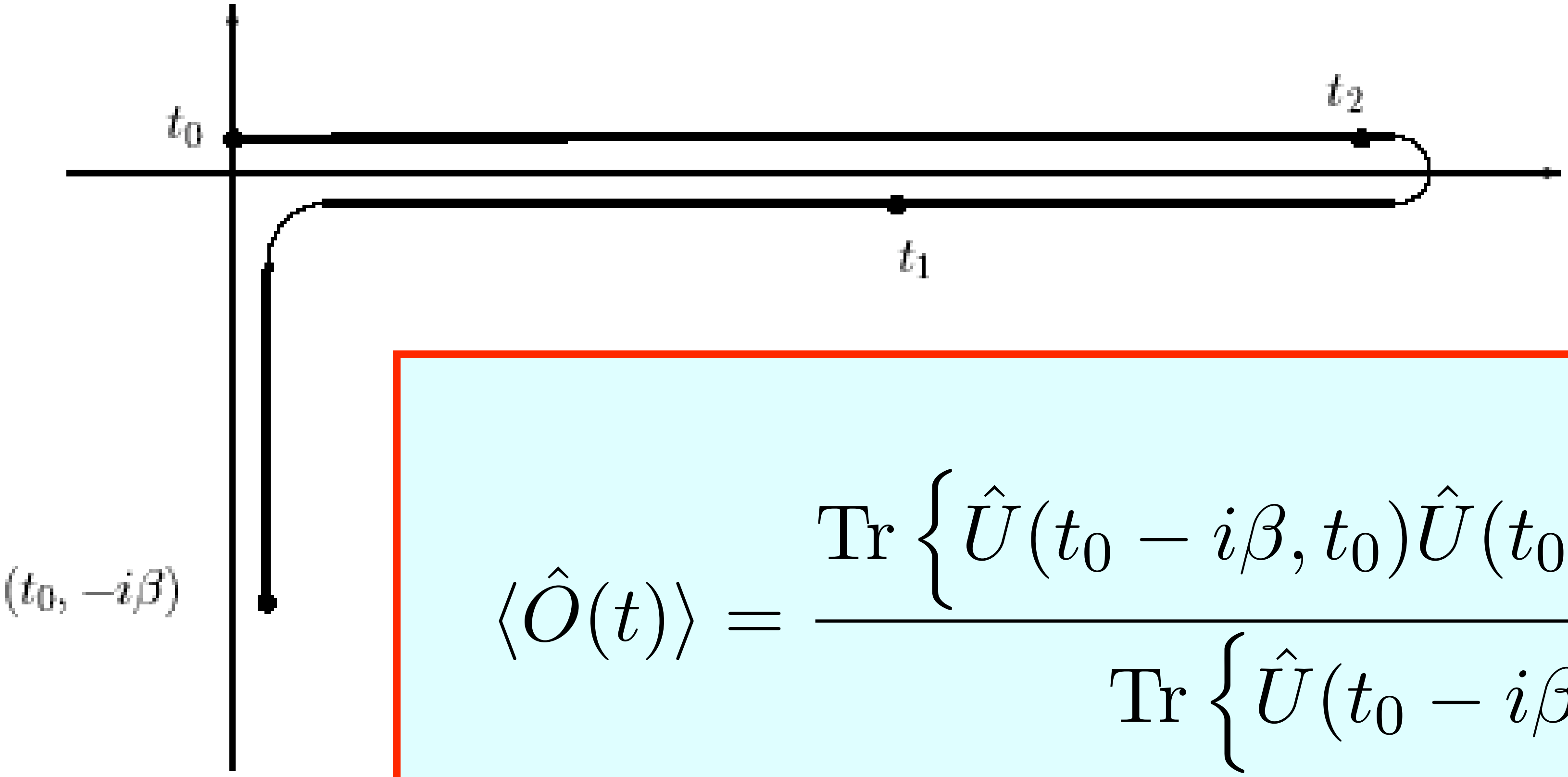
The goal is calculate the time-dependent expectation values of observables :

$$\langle \hat{O}(t) \rangle = \mathrm{Tr}\left\{ \hat{\rho}\, \hat{O}_H(t) \right\}$$

$$\hat{\rho} = \frac{e^{-\beta \hat{H}_0}}{\mathrm{Tr} e^{-\beta \hat{H}_0}}$$

The time contour

(L.V.Keldysh, Sov.Phys.JETP20, 1018 (1965))



$$\langle \hat{O}(t) \rangle = \frac{\mathrm{Tr}\left\{ \hat{U}(t_0 - i\beta, t_0)\hat{U}(t_0, t)\, \hat{O}\, \hat{U}(t, t_0) \right\}}{\mathrm{Tr}\left\{ \hat{U}(t_0 - i\beta, t_0) \right\}}$$

$$\langle \hat{O}(t) \rangle = \frac{\mathrm{Tr}\ \left\{ T_C[\exp(-i \int_C d\bar{t} \hat{H}(\bar{t}) \hat{O}(t)] \right\}}{\mathrm{Tr}\ \left\{ \hat{U}(t_0 - i\beta, t_0) \right\}}$$

Propagators for nonequilibrium systems

We define the Keldysh contour-ordered Green function as :

$$G(1,2) = -i\langle T_C[\hat{\psi}_H(1)\hat{\psi}^\dagger_H(2)]\rangle = \theta(t_1,t_2)G^>(1,2) + \theta(t_2,t_1)G^<(1,2)$$

$$G^>(1,2) = -i\langle\hat{\psi}_H(1)\hat{\psi}^\dagger_H(2)\rangle$$

Propagation of a "particle" (added electron)

$$G^<(1,2) = i\langle\hat{\psi}^\dagger_H(2)\hat{\psi}_H(1)\rangle$$

Propagation of a "hole" (removed electron)

We similarly define the two-particle Green function as :

$$G_2(1,2,3,4) = (-i)^2\langle T_C[\hat{\psi}_H(1)\hat{\psi}_H(2)\hat{\psi}^\dagger_H(3)\hat{\psi}^\dagger_H(4)]\rangle$$

Physical content

The nonequilibrium Green function contains a wealth of detailed information :

- The expectation value of any one-body operator such as electron and current densities, the Wigner distribution function and momentum densities.

- The total energy

- All the electron affinities and ionization energies of the system (photo-electron spectra)

- The excitation energies of the system (absorption spectra)

- Life-times of excitations

The equations of motion

The equations of motion for the Green function are given by :

$$(i\partial_{t_1} - h(1))G(1,1') = \delta(1,1') - i\int d2\, w(1,2)G_2(1,2,2^+,1')$$

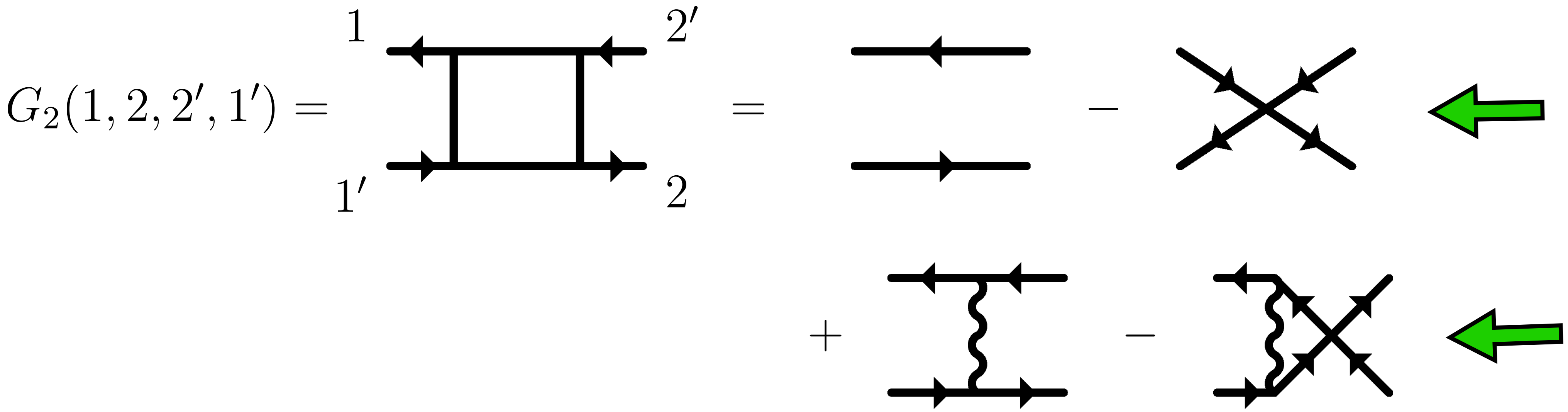
$$(-i\partial_{t_1'} - h(1))G(1,1') = \delta(1,1') - i\int d2\, w(1',2)G_2(1,2,2^+,1')$$

with boundary conditions

$$G(\mathbf{x}_1 t_0 - i\beta, 2) = -G(\mathbf{x}_1 t_0, 2)$$

$$G(1, \mathbf{x}_2 t_0) = -G(1, \mathbf{x}_2 t_0 - i\beta)$$

The simplest collision terms correspond to the following structure of the two-particle Green function:



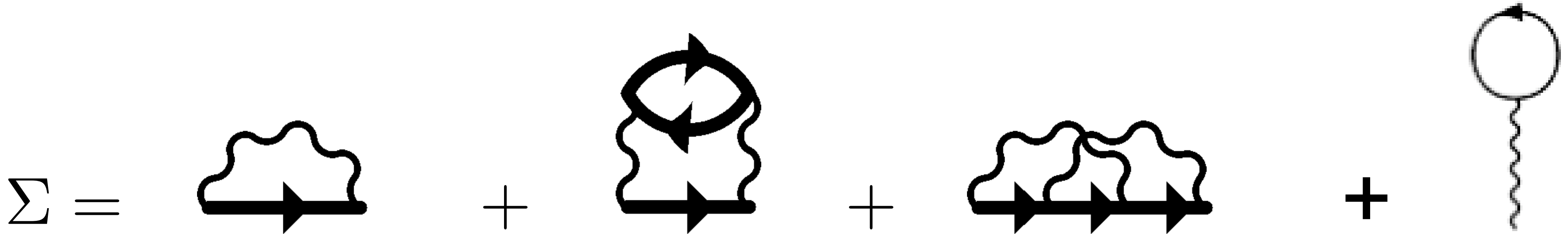
Hartree-Fock terms

collision terms

We define a self-energy operator as follows

$$\int d2\,\Sigma(1,2)G(2,1') = -i\int d2\,w(1,2)\,G_2(1,2,2^+,1')$$

The corresponding self-energy diagrams to 2nd order are :



The equation of motion for the Green function attains the form (these are essentially the Kadanoff-Baym equations):

$$(i\partial_{t_1} - h(1))G(1,2) = \delta(1,2) + \int d3\, \Sigma[G](1,3)G(3,2)$$

A space-time nonlocal potential describing the effects of two-particle interactions

By splitting the equation of motion in components, one obtains the set of Kadanoff-Baym equations. For example for the lesser component $G^<$:

Time-dependent
external field

$$(i\partial_{t_1} - h(1))G^<(1,2) - \int d\mathbf{x}_3 \Sigma^{HF}(1,\mathbf{x}_3 t_1)G^<(\mathbf{x}_3 t_1, 2)$$

$$= \int_{t_0}^{t_1} d3[\Sigma^>(1,3) - \Sigma^<(1,3)]G^<(3,2) - \int_{t_0}^{t_2} d2\,\Sigma^<(1,3)[G^>(3,2) - G^<(3,2)]$$

$$+ \int_{t_0}^{t_0 - i\beta} d3\,\Sigma^\rceil(1,3)G^\lceil(3,2)$$

Collision or electron
correlation terms :
Memory kernels

Initial correlations

The conservation laws

From an approximate Green function we can calculate several observables

$$\langle n(1)\rangle = -iG(1,1^+)$$

density

$$\langle \mathbf{j}(1)\rangle = -i\left[\frac{\nabla_1}{2i} - \frac{\nabla_{1'}}{2i} + \mathbf{A}(1)\right] G(1,1')_{1'=1^+}$$

current density

$$\langle \mathbf{P}(t_1)\rangle = \int d\mathbf{x}_1\, \langle \mathbf{j}(1)\rangle$$

momentum

These observables are related by conservation laws such as

$$\partial_{t_1}\langle n(1)\rangle + \nabla_1 \cdot \langle \mathbf{j}(1)\rangle = 0$$

number conservation

$$\partial_{t_1}\langle \mathbf{P}(t_1)\rangle = -\int d\mathbf{x}_1\, [\langle n(1)\rangle \mathbf{E}(1) + \langle \mathbf{j}(1)\rangle \times \mathbf{B}(1)]$$

momentum conservation

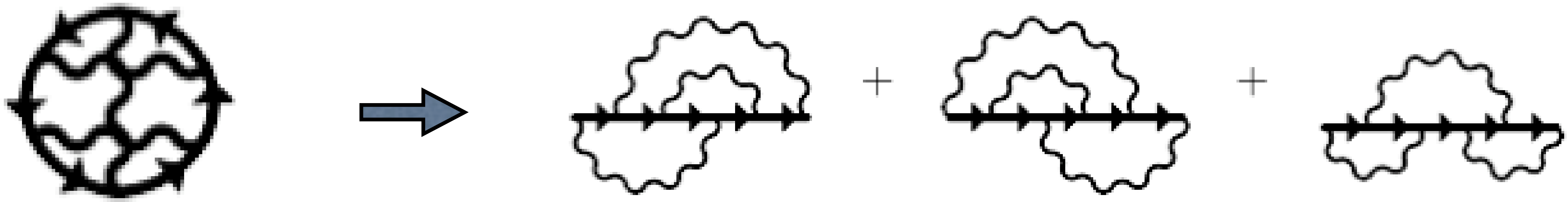
Will these relations be satisfied if the ingredients are calculated from an approximate Green function ?

The conservation laws

(G.Baym, Phys.Rev. 127, 1391 (1962))

Conservation laws, such as those of energy, momentum, angular momentum and particle number, are automatically obeyed when we use so-called Phi-derivable approximations for the self-energy.

$$\boxed{\Sigma(1,2) = \frac{\delta\Phi}{\delta G(2,1)}}$$



For Phi-derivable approximations the expectation values are independent from the way they are calculated

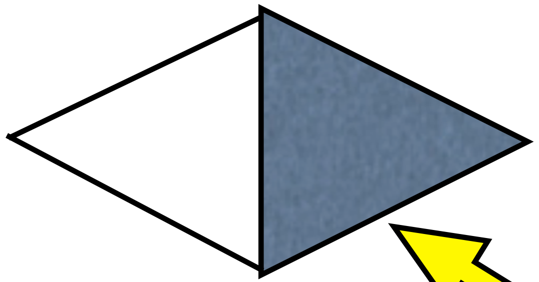
f-sum rule and Ward identity

The f-sumrule and the Ward identity are satisfied for the response function

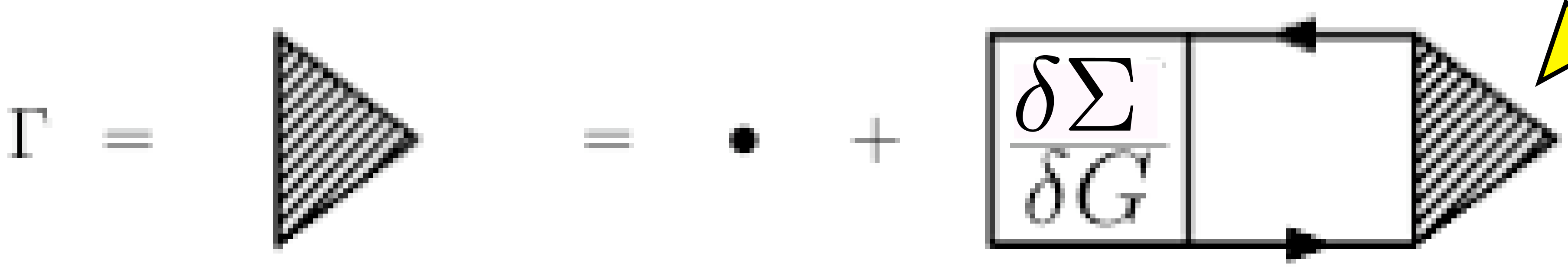
$$\delta n(1) = -i\delta G(1,1^+) = \int d2\, \chi(1,2)\delta v(2)$$

where

$$\chi = P + Pv\chi \qquad P = GG\Gamma =$$



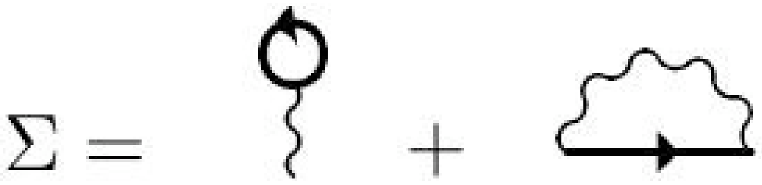
vertex



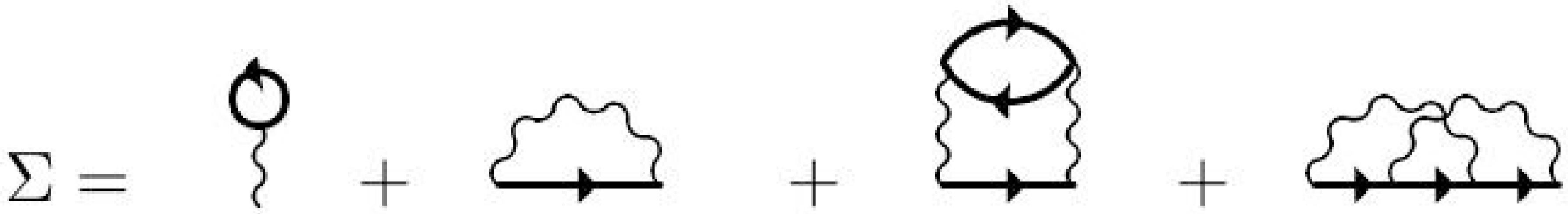
see e.g. N.H.Kwong, M.Bonitz, Phys.Rev.Lett. 84, 1768 (2000)

Conserving many-body approximations

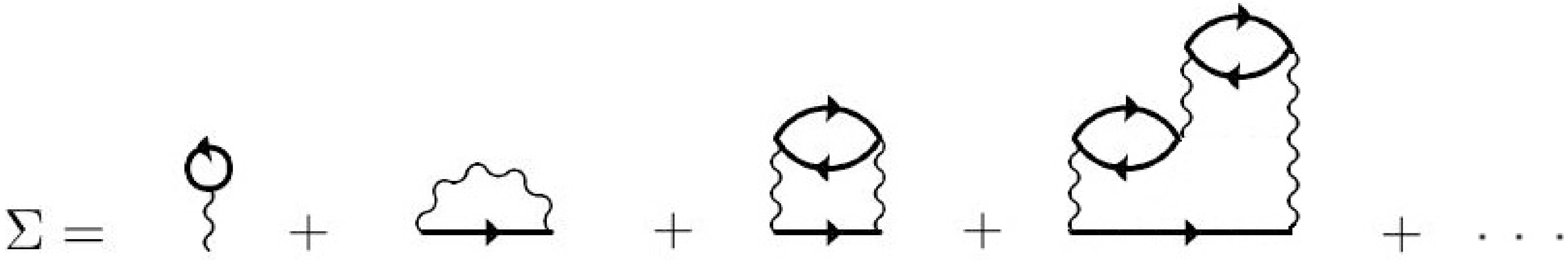
Hartree-Fock



2nd Born



GW



Practical solution of the equations of motion

For practical solution the Green function is expanded into one-particle states

$$G(1,2) = \sum_{ij} \varphi_i(\mathbf{x}_1) G_{ij}(t_1,t_2) \varphi_j^*(\mathbf{x}_2)$$

$$G_{ij}(t_1,t_2) = -i \langle T_C \hat{a}_{i,H}(t_1) \hat{a}^\dagger_{j,H}(t_2) \rangle$$

For the one-particle states we can, for instance, use the solutions to the Hartree-Fock or Kohn-Sham equations

The Kadanoff-Baym equations become equations for time-dependent matrices

To evaluate the many-body interactions in the self-energy we calculate

$$v_{ijkl} = \int d\mathbf{x} \int d\mathbf{x}' \, \varphi_i^*(\mathbf{x})\varphi_j^*(\mathbf{x}')v(\mathbf{r}-\mathbf{r}')\varphi_k(\mathbf{x}')\varphi_l(\mathbf{x})$$

The self-energy for second Born is e.g. given by

$$\Sigma^{2B}(t,t') = \delta(t,t')\Sigma^{HF}(t) + \Sigma^{(2)}(t,t')$$

$$\Sigma_{ij}^{HF}(t) = -i\sum_{kl} G_{kl}(t,t^+)(2v_{ilkj} - v_{iljk})$$

$$\Sigma_{ij}^{(2)}(t,t') = \sum_{klmnpq} G_{kl}(t,t')G_{mn}(t,t')G_{pq}(t',t)\, v_{iqmk}(2v_{lnpj} - v_{nlpj})$$

(Nils Erik Dahlen, RvL,
Phys.Rev.Lett. 98, 153004 (2007))

The number of two-electron integrals v_ijkl grows as N^4 . In a localized basis we may use distance effects. The simplest is:

$$v_{ijkl} \approx \delta_{jk}\delta_{il}V_{ij}$$

$$V_{ij} = \int d\mathbf{x} \int d\mathbf{x}' v(\mathbf{r}-\mathbf{r}')|\varphi_i(\mathbf{x})|^2|\varphi_j(\mathbf{x}')|^2$$

The second Born self-energy is then considerably simplified to

$$\Sigma^{(2)}_{ij}(t,t') = 2G_{ij}(t,t')\sum_{pq} G_{pq}(t,t')G_{qp}(t',t)V_{iq}V_{qj} - \sum_{pq} G_{iq}(t,t')G_{qp}(t',t)G_{pj}(t,t')V_{ip}V_{qj}$$

GW and the product basis technique

The GW approximation involves the dynamically screened interaction

$$W = v + vPW \qquad\qquad P = -iGG$$

If we consider the time nonlocal part of W

$$\tilde{W} = W - v$$

we have to solve the equation

$$\tilde{W}_{pqrs}(t,t') = \sum_{ijkl} v_{plis} P_{ijkl}(t,t') v_{jqrk} + \sum_{ijkl} \int dt_1 \, v_{plis} P_{ijkl}(t,t_1) \tilde{W}_{jqrk}(t_1,t')$$

This equation is of the form

$$\tilde{W}_{Q_1Q_2}(t,t') = \sum_{Q_3Q_4} v_{Q_1Q_3} P_{Q_3Q_4}(t,t') v_{Q_4Q_2} + \sum_{Q_3Q_4} \int dt_1\, v_{Q_1Q_3} P_{Q_3Q_4}(t,t_1) \tilde{W}_{Q_4Q_2}(t_1,t')$$

in terms of the nonorthogonal basis

$$f_Q(\mathbf{r}) = \phi_i(\mathbf{r})\phi_j^*(\mathbf{r}) \qquad\qquad Q=(ij)$$

We can reduce the size of the basis considerable by taking only by using a new orthogonal basis

$$g_q(\mathbf{r}) = \frac{1}{\sqrt{\sigma_q}} \sum_{q'} U_{q'q} f_{q'}(\mathbf{r})$$

$$\sum_{q_1q_2} U^\dagger_{qq_1} \langle f_{q_1} | f_{q_2} \rangle U_{q_2q'} = \sigma_q \delta_{qq'}$$

and discarding all basis functions for which $\sigma_q < \epsilon$

(A.Stan,N.E.Dahlen, RvL J.Chem.Phys. 130,114105 (2009))

The Kadanoff-Baym equations

(Nils Erik Dahlen, RvL,
Phys.Rev.Lett. 98, 153004 (2007))

$$\begin{aligned} i\partial_t G^{\lessgtr}(t;t') &= h(t)G^{\lessgtr}(t;t') + I^{\lessgtr}(t;t') \qquad \text{(+ adjoint)} \\ i\partial_t G^{\rceil}(t;i\tau) &= h(t)G^{\rceil}(t;i\tau) + I^{\rceil}(t;i\tau) \\ -i\partial_t G^{\lceil}(i\tau,t) &= G^{\lceil}(i\tau,t)h(t) + I^{\lceil}(i\tau,t) \end{aligned}$$

with the initial conditions

$$G^{<}(0,0) = iG^{M}(0^{-}) \quad , \quad G^{>}(0,0) = iG^{M}(0^{+})$$

$$G^{\rceil}(t,i\tau) = iG^{M}(-i\tau) \quad , \quad G^{\lceil}(i\tau,t) = iG^{M}(i\tau)$$

The collision integrals are

$$\begin{aligned} I^{\lessgtr}(t,t') &= \int_0^{\infty} d\bar{t}\,\left[\Sigma_c^R(t,\bar{t})G^{\lessgtr}(\bar{t},t') + \Sigma_c^{\lessgtr}(t,\bar{t})G^{A}(\bar{t},t')\right] \\ &\quad + \frac{1}{i}\int_0^{-\beta} d\bar{\tau}\,\Sigma^{\rceil}(t,i\bar{\tau})G^{\lceil}(i\bar{\tau},t') \\ I^{\rceil}(t,i\tau') &= \int_0^{\infty} d\bar{t}\,\Sigma_c^R(t,\bar{t})G^{\rceil}(\bar{t},i\tau') \\ &\quad + \int_0^{-\beta} d\bar{\tau}\,\Sigma^{\rceil}(t,i\bar{\tau})G^{M}(i(\bar{\tau}-\tau')) \end{aligned}$$

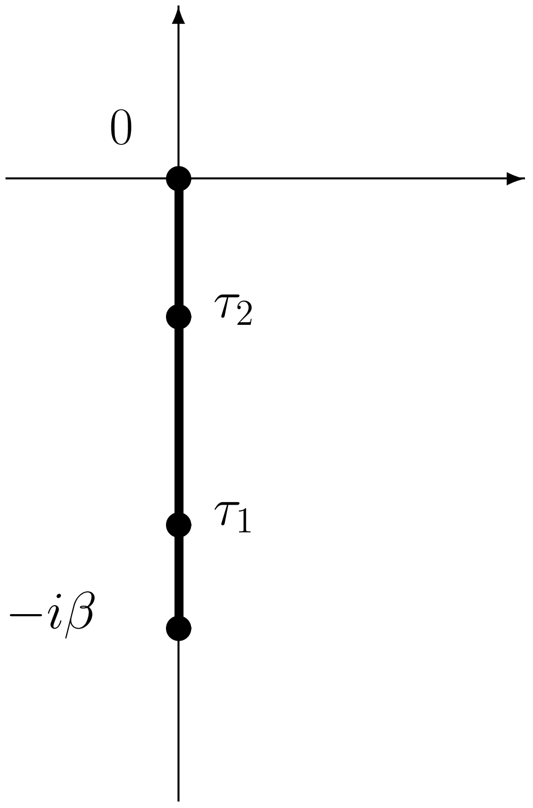
and $[I^{\lceil}(i\tau,t)]^{\dagger} = I^{\rceil}(t,i\tau)$.

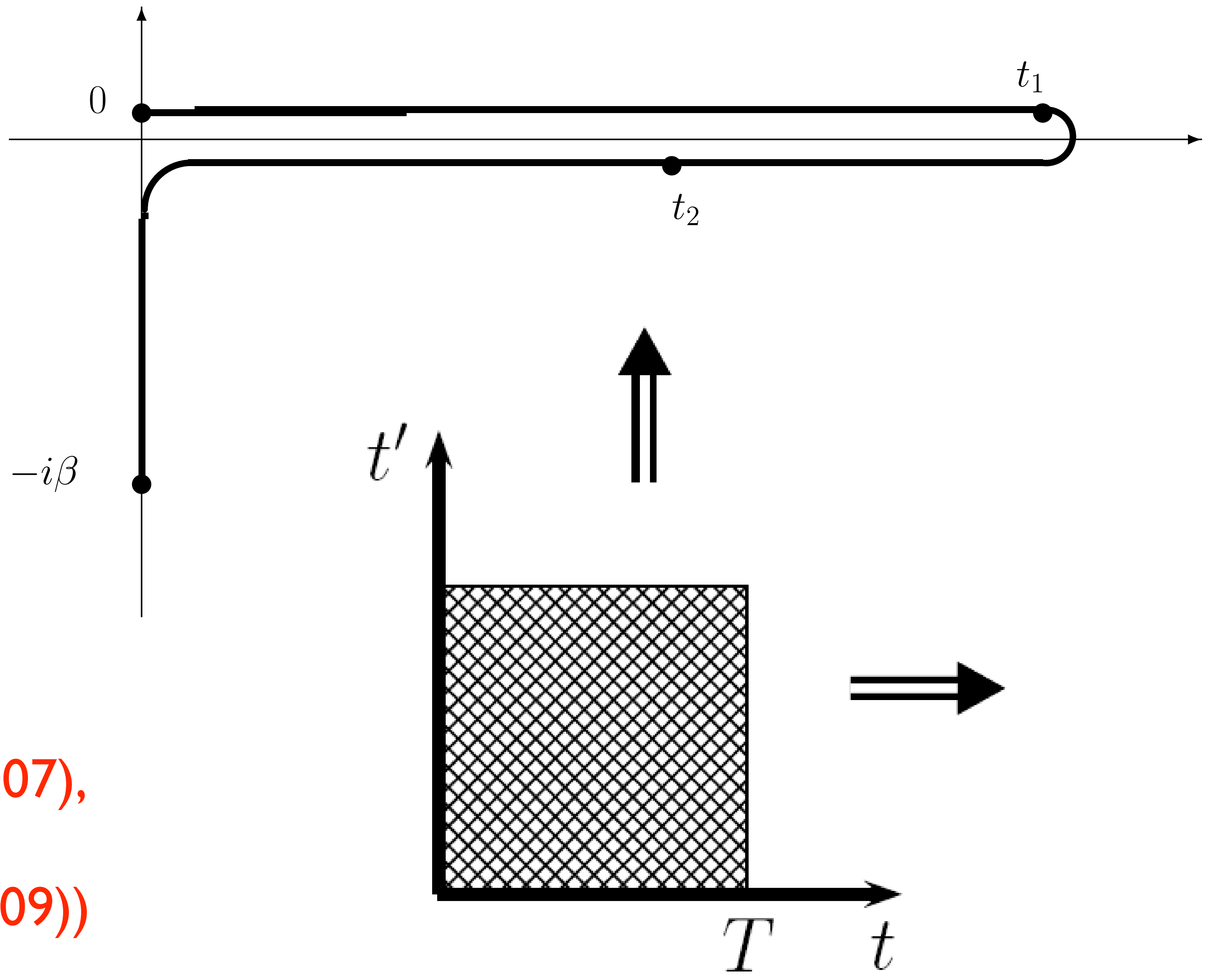
Splitting the equation of motion into components on different parts of the Keldysh contour

Time propagation of the Kadanoff-Baym equations

Solve equilibrium case on the imaginary axis

Carry out time-stepping in the double-time plane (possibly with external field applied)





(Nils Erik Dahlen, RvL,
Phys.Rev.Lett. 98, 153004 (2007),
A.Stan, N.E.Dahlen, RvL,
J.Chem.Phys.130, 224101 (2009))

The way we solve the equations of motion requires the storage of the matrix elements G_{KL} (t,t’) for time-points t and t’, which grows quadratically with the number of basis functions and with the number of time points.

I would be a great advantage to improve the scaling of the time-propagation scheme:

T^2 → $T \ln T$ -scaling ?

See: M.Zwolak,
“Numerical ansatz for solving integro-differential equations with increasingly smooth memory kernels¨

Computational Science and Discovery 1, 015002 (2008)

The hydrogen molecule in a laser field

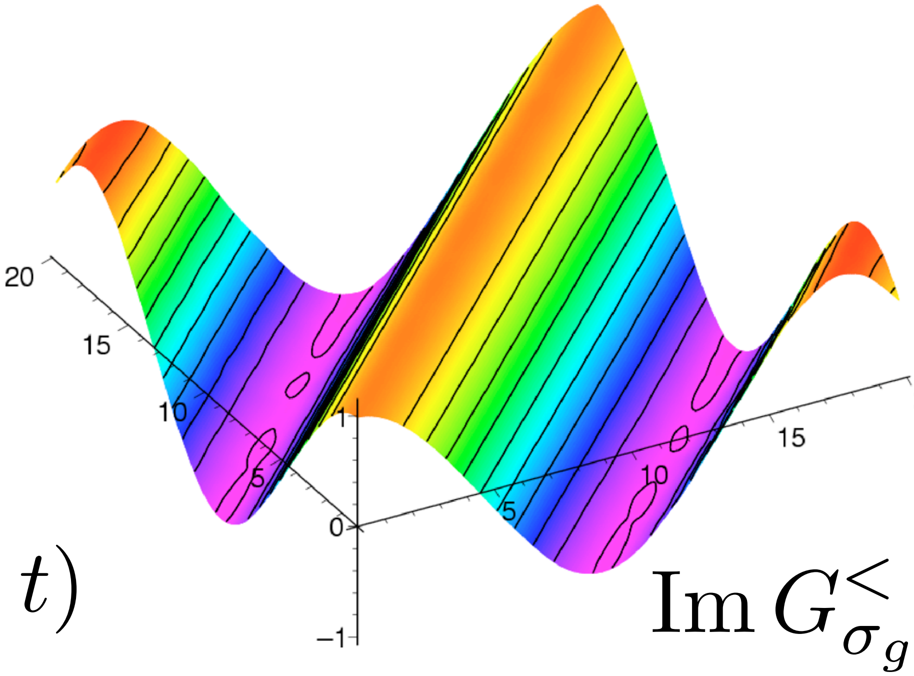
$$v(\mathbf{r}t) = E(t)z$$

$$E(t) = \theta(t - t_0)E_0$$

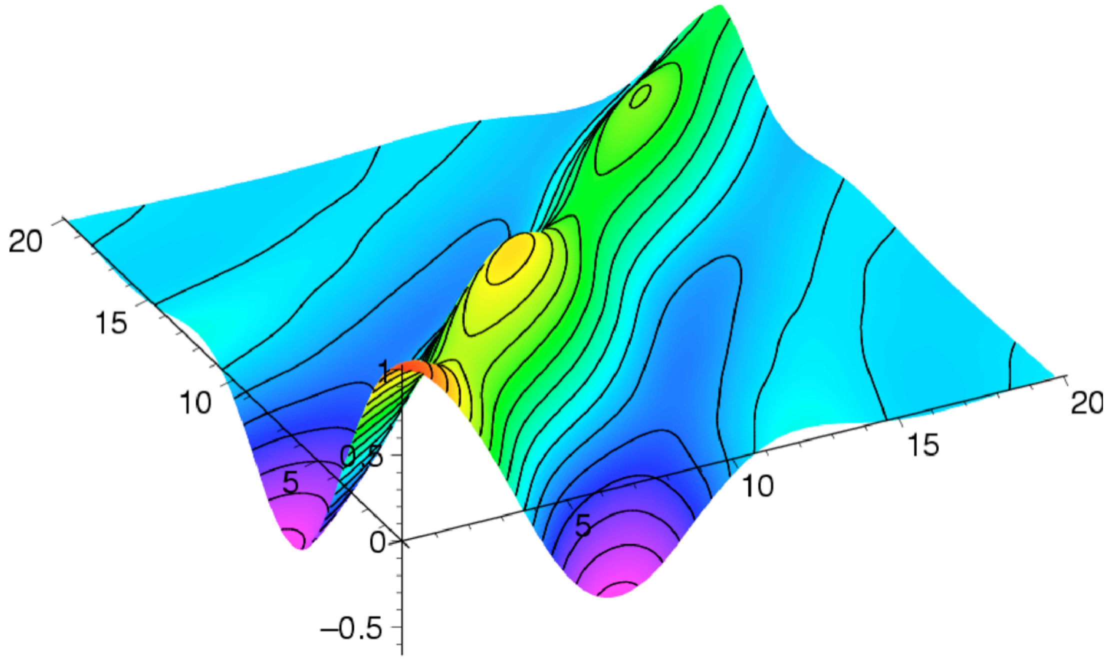
On the time diagonal :

$$n_i(t) = \langle \hat{a}^\dagger_{i,H}(t)\hat{a}_{i,H}(t)\rangle = \mathrm{Im}\, G^<_{ii}(t,t)$$

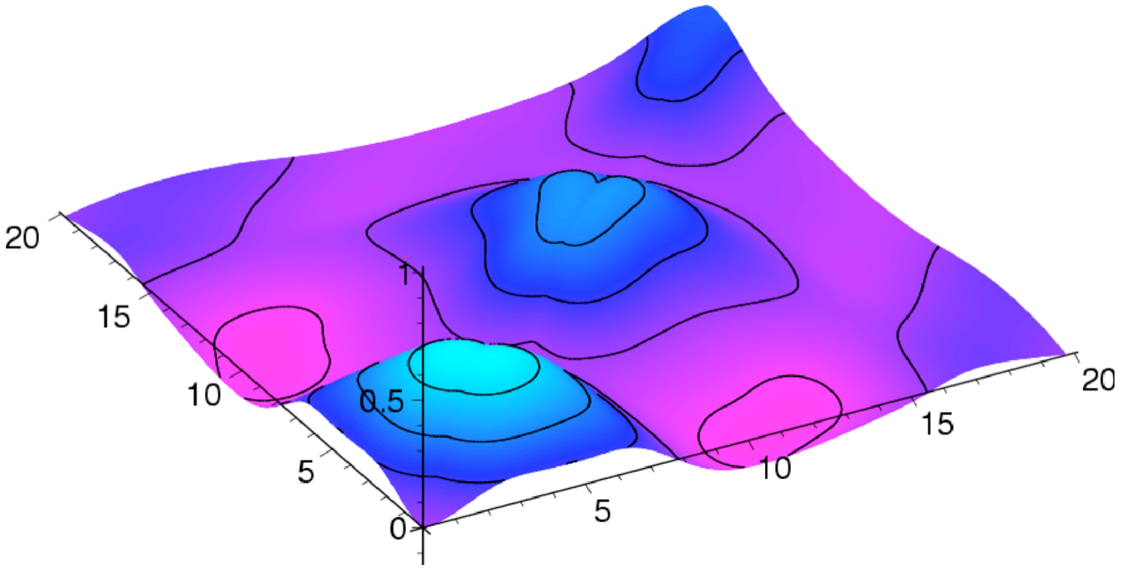
Equilibrium (no field applied)



$$\mathrm{Im}\, G^<_{\sigma_g\sigma_g}(t_1,t_2)$$



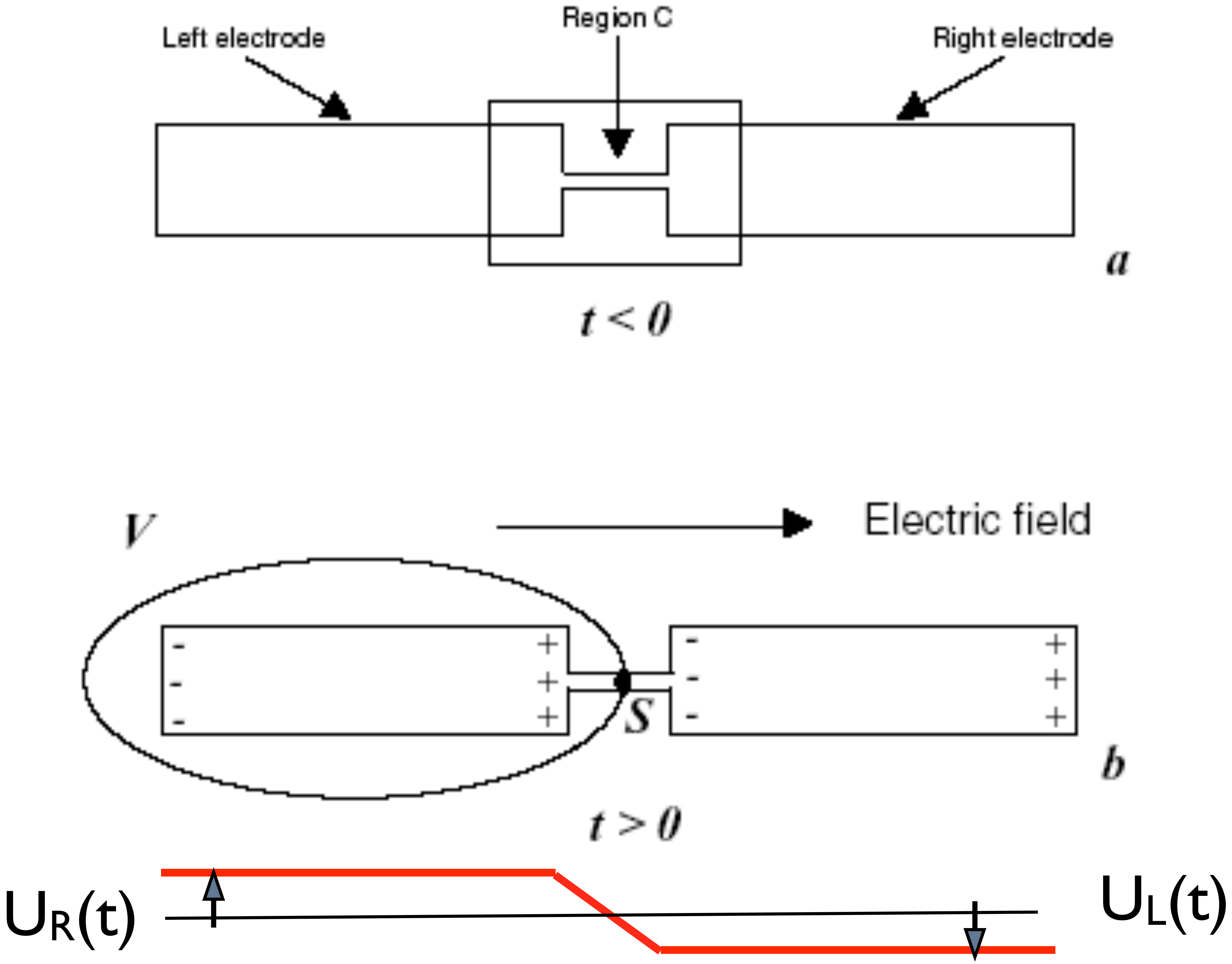
$$\mathrm{Im}\, G^<_{\sigma_g\sigma_g}(t_1,t_2)$$



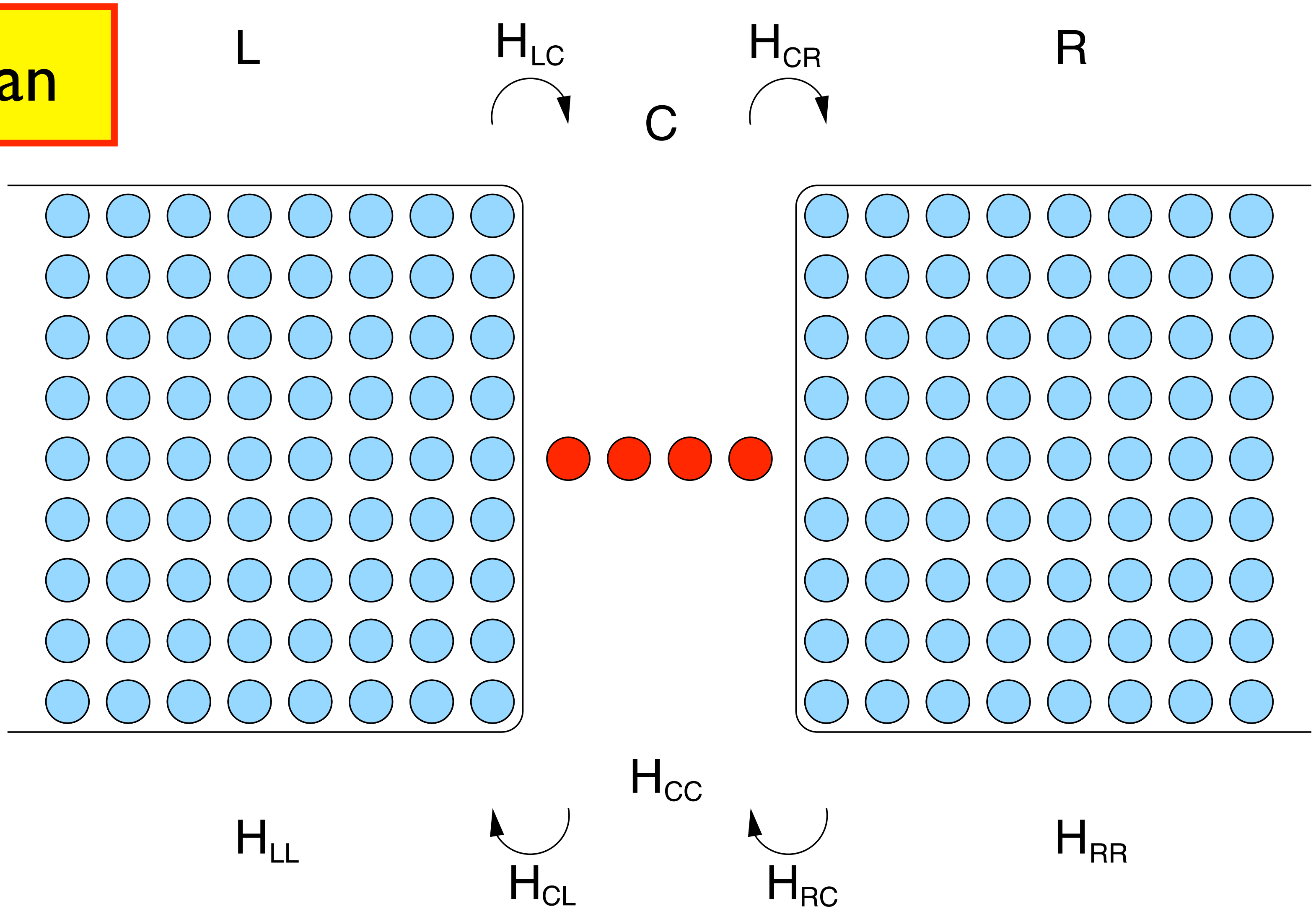
$$\mathrm{Im}\, G^<_{\sigma_u\sigma_u}(t_1,t_2)$$

Nonequilibrium (field applied)

The quantum conduction problem



Model Hamiltonian



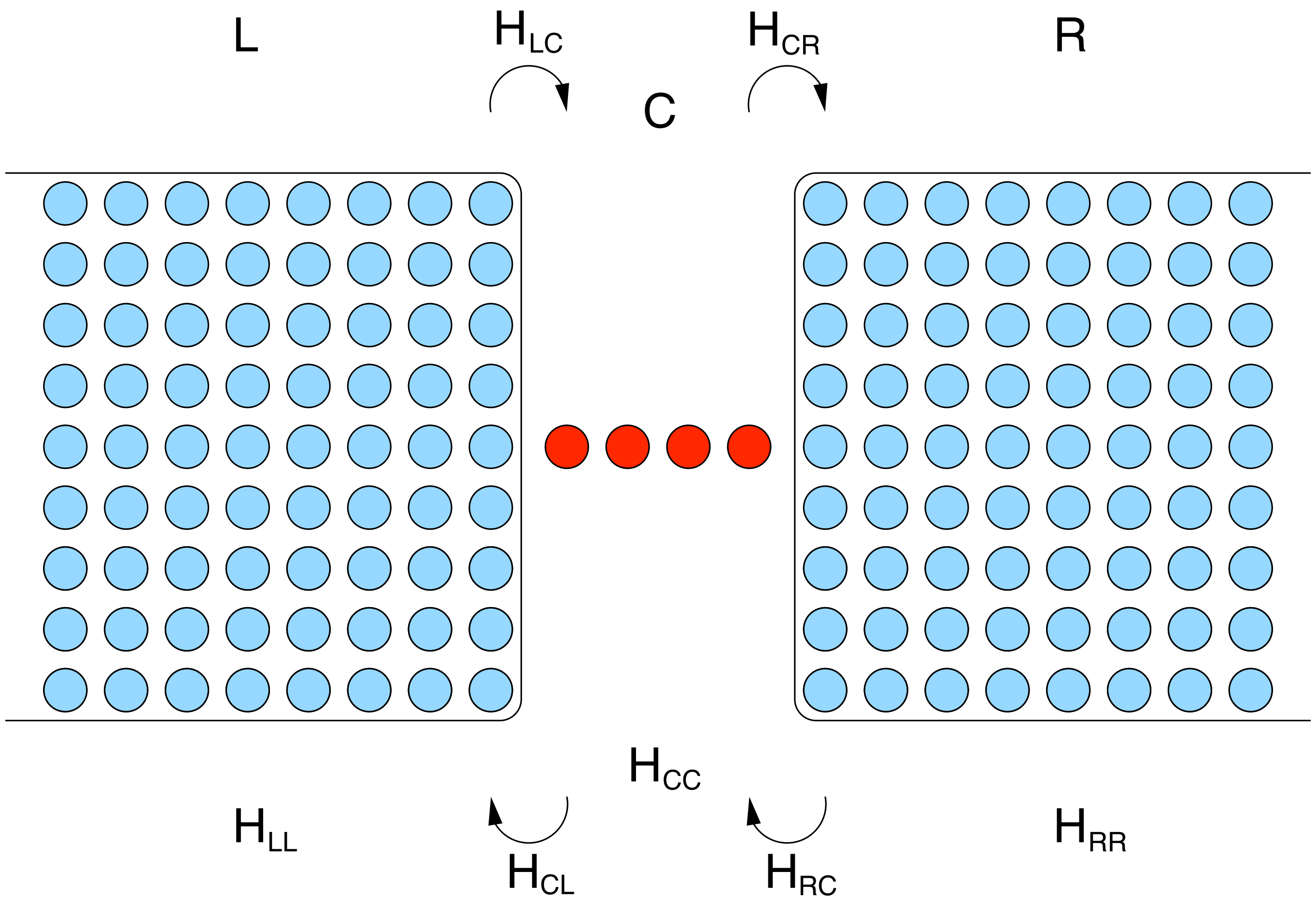
$$\hat{H}_C(t) = \sum_{ij,\sigma} h_{ij}(t)\hat{d}^\dagger_{i\sigma}\hat{d}_{j\sigma} + \frac{1}{2}\sum_{\substack{ijkl\\ \sigma\sigma'}} v_{ijkl}\hat{d}^\dagger_{i\sigma}\hat{d}^\dagger_{j\sigma'}\hat{d}_{k\sigma'}\hat{d}_{l\sigma}$$

$$\hat{H}_\alpha(t) = U_\alpha(t)\hat{N}_\alpha + \sum_{ij,\sigma} h^\alpha_{ij}\,\hat{c}^\dagger_{i\sigma\alpha}\hat{c}_{j\sigma\alpha}$$

$$\hat{H}_T = \sum_{ij,\sigma\alpha} V_{i,j\alpha}[\hat{d}^\dagger_{i\sigma}\hat{c}_{j\sigma\alpha} + \hat{c}^\dagger_{j\sigma\alpha}\hat{d}_{i\sigma}]$$

The embedding

The one-body part of the Hamiltonian is projected onto different regions



$$\mathbf{H} = \begin{bmatrix} \mathbf{H}_{\mathrm{LL}} & \mathbf{H}_{\mathrm{LC}} & \mathbf{0} \\ \mathbf{H}_{\mathrm{CL}} & \mathbf{H}_{\mathrm{CC}} & \mathbf{H}_{\mathrm{CR}} \\ \mathbf{0} & \mathbf{H}_{\mathrm{RC}} & \mathbf{H}_{\mathrm{RR}} \end{bmatrix}$$

The Green function and the self-energy attain the form

$$\boldsymbol{\mathcal{G}} = \begin{bmatrix} \boldsymbol{\mathcal{G}}_{\mathrm{LL}} & \boldsymbol{\mathcal{G}}_{\mathrm{LC}} & \boldsymbol{\mathcal{G}}_{\mathrm{LR}} \\ \boldsymbol{\mathcal{G}}_{\mathrm{CL}} & \boldsymbol{\mathcal{G}}_{\mathrm{CC}} & \boldsymbol{\mathcal{G}}_{\mathrm{RC}} \\ \boldsymbol{\mathcal{G}}_{\mathrm{RL}} & \boldsymbol{\mathcal{G}}_{\mathrm{CR}} & \boldsymbol{\mathcal{G}}_{\mathrm{RR}} \end{bmatrix} \qquad \boldsymbol{\Sigma}^{\mathrm{MB}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & \boldsymbol{\Sigma}^{\mathrm{MB}}_{\mathrm{CC}}[\boldsymbol{\mathcal{G}}_{\mathrm{CC}}] & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

with equations of motion for the complete system

$$\begin{aligned} i\partial_z \boldsymbol{\mathcal{G}}(z,z') &= \delta(z,z')\mathbf{1} + \mathbf{H}(z)\boldsymbol{\mathcal{G}}(z,z') \\ &+ \int d\bar{z}\, \boldsymbol{\Sigma}^{\mathrm{MB}}(z,\bar{z})\boldsymbol{\mathcal{G}}(\bar{z},z') \\ -i\partial_{z'} \boldsymbol{\mathcal{G}}(z,z') &= \delta(z,z')\mathbf{1} + \boldsymbol{\mathcal{G}}(z,z')\mathbf{H}(z') \\ &+ \int d\bar{z}\, \boldsymbol{\mathcal{G}}(z,\bar{z})\boldsymbol{\Sigma}^{\mathrm{MB}}(\bar{z},z) \end{aligned}$$

The projection on region CC gives

$$\Big\{ i\partial_z \mathbf{1} - \mathbf{H}_{\mathrm{CC}}(z) \Big\} \boldsymbol{\mathcal{G}}_{\mathrm{CC}}(z,z') = \delta(z,z')\mathbf{1} + \\ \sum_{\alpha} \mathbf{H}_{\mathrm{C}\alpha} \boldsymbol{\mathcal{G}}_{\alpha\mathrm{C}}(z,z') + \int d\bar{z}\, \boldsymbol{\Sigma}^{\mathrm{MB}}_{\mathrm{CC}}(z,\bar{z}) \boldsymbol{\mathcal{G}}_{\mathrm{CC}}(\bar{z},z')$$

while the projection on region aC gives

$$\Big\{ i\partial_z \mathbf{1} - \mathbf{H}_{\alpha\alpha}(z) \Big\} \boldsymbol{\mathcal{G}}_{\alpha\mathrm{C}}(z,z') = \mathbf{H}_{\alpha\mathrm{C}} \boldsymbol{\mathcal{G}}_{\mathrm{CC}}(z,z')$$

This can be solved to give

$$\boldsymbol{\mathcal{G}}_{\alpha\mathrm{C}}(z,z') = \int d\bar{z}\, \mathbf{g}_{\alpha\alpha}(z,\bar{z})\, \mathbf{H}_{\alpha\mathrm{C}} \boldsymbol{\mathcal{G}}_{\mathrm{CC}}(\bar{z},z')$$

where the biased but uncontacted lead Green function satisfies

$$\Big\{ i\partial_z \mathbf{1} - \mathbf{H}_{\alpha\alpha}(z) \Big\} \boldsymbol{g}_{\alpha\alpha}(z,z') = \delta(z,z')\mathbf{1}$$

The equation of motion

The equation of motion projected on the central region has the form

$$\left\{ i\partial_z \mathbf{1} - \mathbf{H}_{\mathrm{CC}}(z) \right\} \boldsymbol{\mathcal{G}}_{\mathrm{CC}}(z,z')$$
$$= \delta(z,z')\mathbf{1} + \int d\bar{z} \left[\boldsymbol{\Sigma}^{\mathrm{MB}}_{\mathrm{CC}} + \boldsymbol{\Sigma}_{\mathrm{em}} \right] (z,\bar{z})\, \boldsymbol{\mathcal{G}}_{\mathrm{CC}}(\bar{z},z').$$

where on top of the a many-body self-energy we also have
an effective embedding self-energy

$$\boldsymbol{\Sigma}_{\mathrm{em}}(z,z') = \sum_{\alpha} \boldsymbol{\Sigma}_{\mathrm{em},\alpha}(z,z') = \sum_{\alpha} \mathbf{H}_{\mathrm{C}\alpha}\, \mathbf{g}_{\alpha\alpha}(z,z') \mathbf{H}_{\alpha\mathrm{C}}$$

Numerical aspects:

- Embedding self-energy independent of G_{CC}

- Now even the time-dependent Hartree-Fock equations involve memory due to embedding.

- When interactions in the leads need to be taken into account the central region may become big.

Calculating the current

The total current flowing out of reservoir α is given by :

$$I_\alpha(t) = \frac{dN_\alpha(t)}{dt} = -2\,\mathrm{Re}\mathrm{Tr}_C[G^<_{C\alpha}(t,t)H_{\alpha C}]$$

This gives after some manipulations:

$$\begin{aligned} I_\alpha(t) = &-2\mathrm{ReTr}_C \int_0^t dt' \left[G^<_{CC}(t,t')\Sigma^{\mathrm{A}}_{\mathrm{em},\alpha}(t',t) + G^{\mathrm{R}}_{CC}(t,t')\Sigma^<_{\mathrm{em},\alpha}(t,t')\right] \\ &-2\mathrm{ReTr}_C \int_0^{-i\beta} dt' \left[G^\rceil_{CC}(t,t')\Sigma^\lceil_{\mathrm{em},\alpha}(t',t)\right] \end{aligned}$$

Memory of initial correlations

Long time limit leads under some assumptions to Meir-Wingreen formula

The spectral function

The spectral function for a nonequilibrium system is defined as

$$A(t,t') = \mathrm{Tr}\,\mathbf{A}(t,t') \qquad \mathbf{A}_{ij}(t,t') = \langle \Psi_0 | \{ \hat{a}_{i,H}(t), \hat{a}^\dagger_{j,H}(t') \} | \Psi_0 \rangle$$

In equilibrium the spectral function only depends on the difference of the time coordinates and can be Fourier transformed to give

$$\begin{aligned} A_{ii}(\omega) = & \sum_k |\langle \Psi_k^{N+1} | \hat{a}_i^\dagger | \Psi_0 \rangle|^2 \delta(\omega + E_0^N - E_k^{N+1}) \\ & + \sum_k |\langle \Psi_k^{N-1} | \hat{a}_i | \Psi_0 \rangle|^2 \delta(\omega - E_0^N + E_k^{N-1}) \end{aligned}$$

It shows peaks at electron addition and removal energies

In the nonequilibrium case it is convenient to Fourier transform with respect to the relative times:

$$A(T,\omega) = \int \frac{d\omega}{2\pi} A(T+\frac{t}{2}, T-\frac{t}{2})\, e^{i\omega t}$$

which can be calculated from the Green function as

$$\boxed{A(T,\omega) = -\mathrm{Im}\mathrm{Tr}_{\mathrm{C}} \int \frac{dt}{2\pi}\, e^{i\omega t} [\boldsymbol{\mathcal{G}}^{>}_{\mathrm{CC}} - \boldsymbol{\mathcal{G}}^{<}_{\mathrm{CC}}](T+\frac{t}{2}, T-\frac{t}{2})}$$

In the long time limit the spectral function becomes independent of T when a steady state is being reached

$$\lim_{T\to\infty} A(T,\omega) = A(\omega)$$

Density in the leads

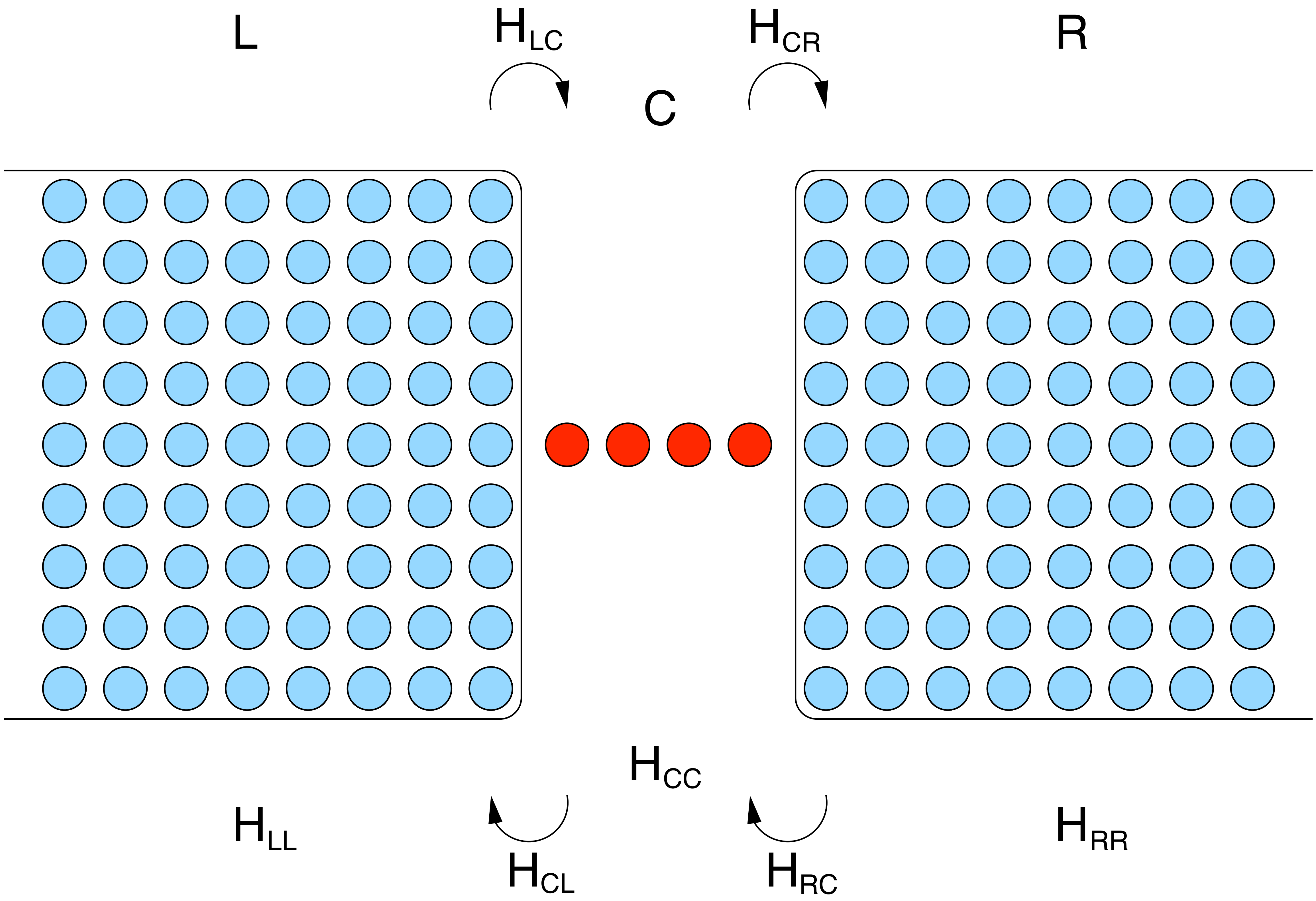
If we define the inbedding self-energy as

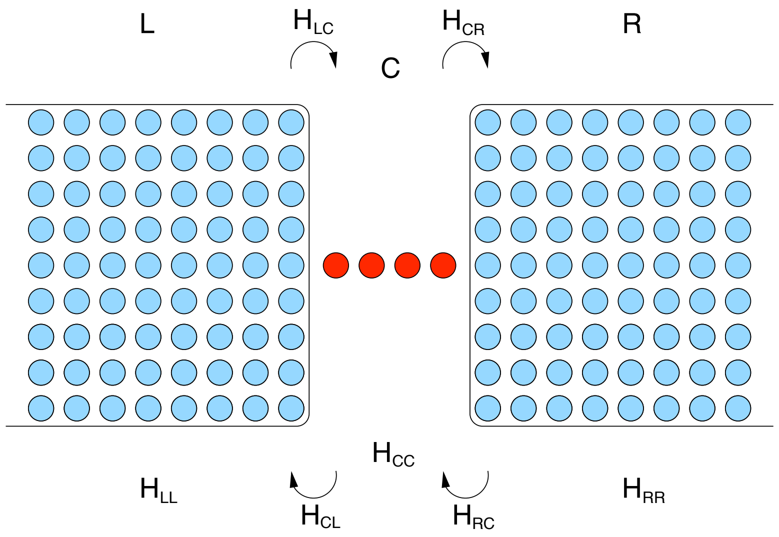
$$\mathbf{\Sigma}_{\mathrm{in},\alpha}(z,z') = \mathbf{H}_{\alpha\mathrm{C}}\boldsymbol{\mathcal{G}}_{\mathrm{CC}}(z,z')\mathbf{H}_{\mathrm{C}\alpha}$$

Then the densities in the leads can be calculated from the equation

$$\begin{aligned}\boldsymbol{\mathcal{G}}_{\alpha\alpha}(t_-,t_+) &= \boldsymbol{g}_{\alpha\alpha}(t_-,t_+) + \\ &+ \int d\bar{z}d\bar{\bar{z}}\boldsymbol{g}_{\alpha\alpha}(t_-,\bar{z})\mathbf{\Sigma}_{\mathrm{in},\alpha}(\bar{z},\bar{\bar{z}})\boldsymbol{g}_{\alpha\alpha}(\bar{\bar{z}},t_+)\end{aligned}$$

Results: 4 atom chain connected to 9-row two-dimensional leads



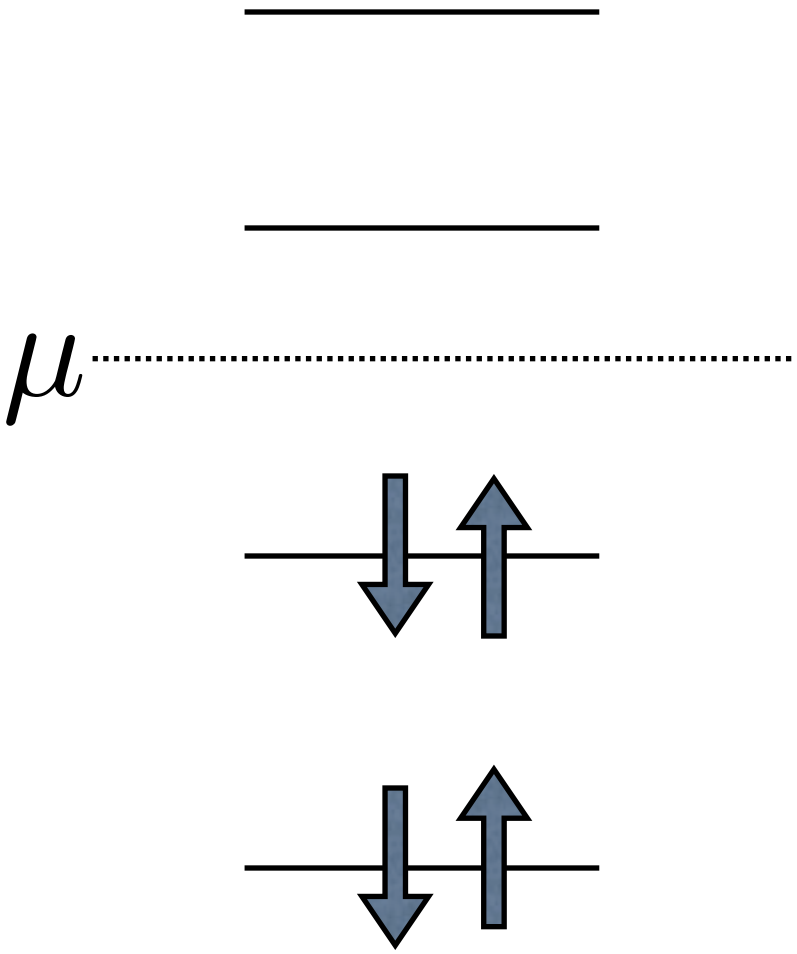


Interaction

$$v_{ijkl} = v_{ij}\delta_{il}\delta_{jk}$$

$$v_{ij} = \begin{cases} v_{ii} & i = j \\ \frac{v_{ii}}{2|i-j|} & i \neq j \end{cases}$$

$$v_{ii} = 1.5$$



Time-dependent bias

$$U_L(t) = -U_R(t) = U\,\theta(t-t_0)$$

Hoppings:

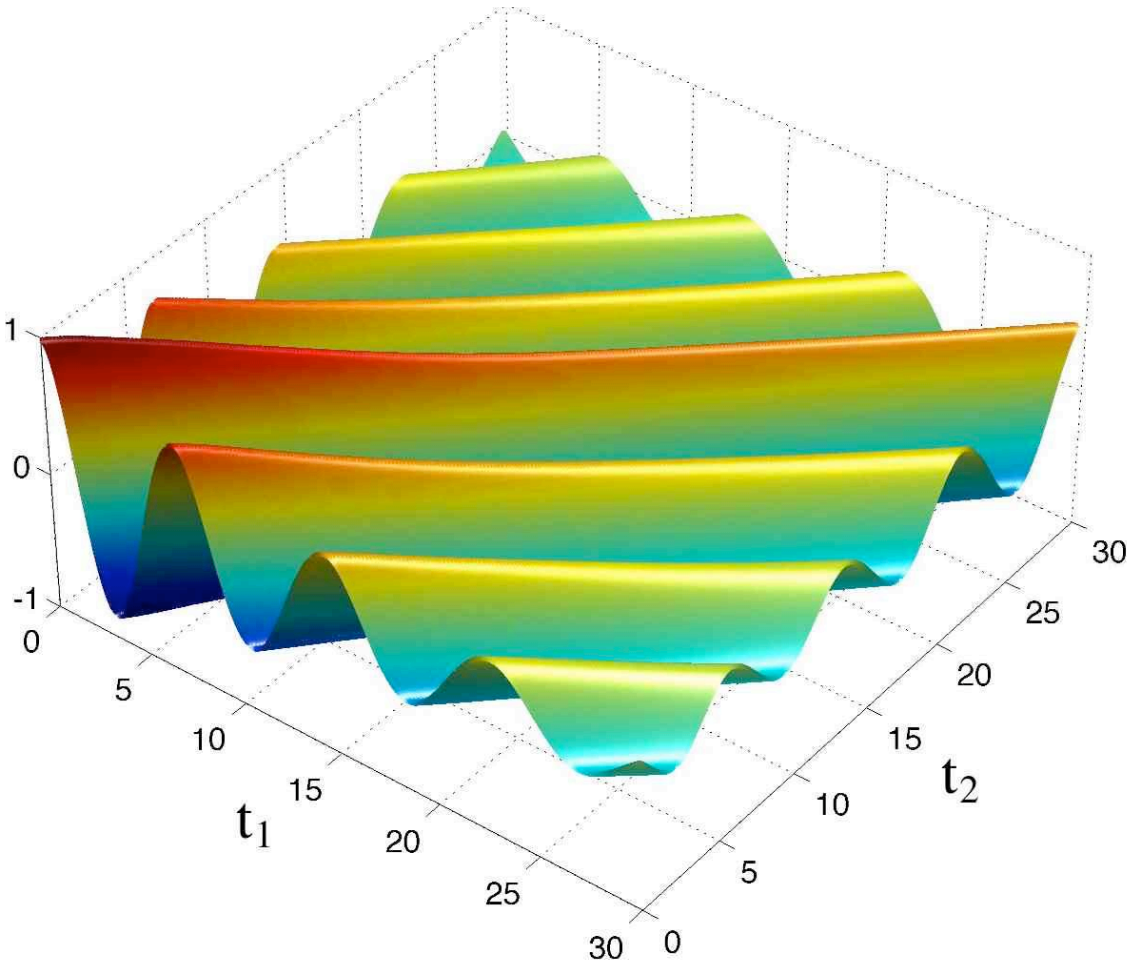
$$t^\alpha = -2$$

$$t_C = -1$$

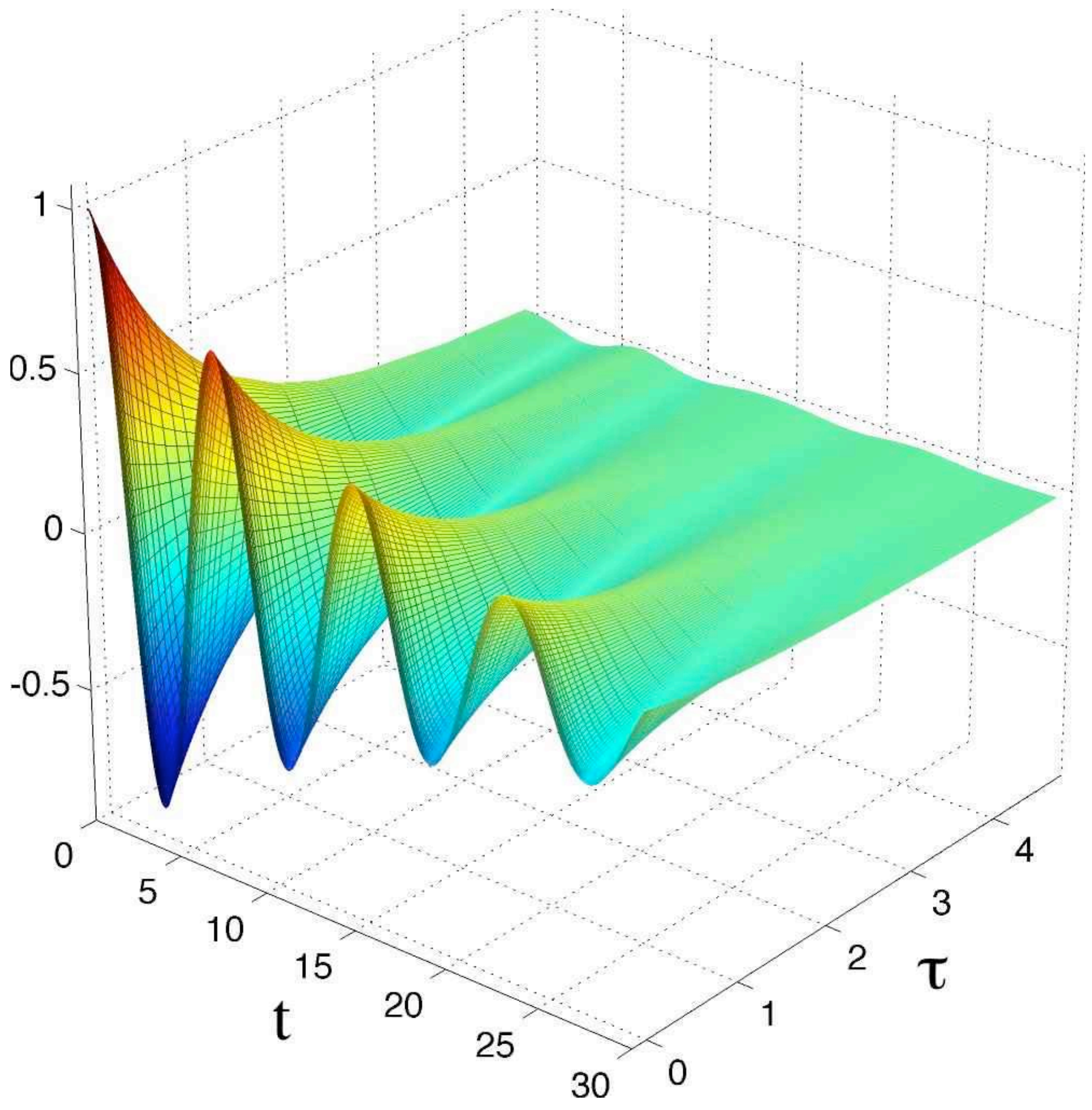
$$V_{1,5L} = V_{4,5R} = -0.5$$

The Green function

For the highest occupied molecular orbital the Green function matrix element has the following structure (imaginary part displayed)

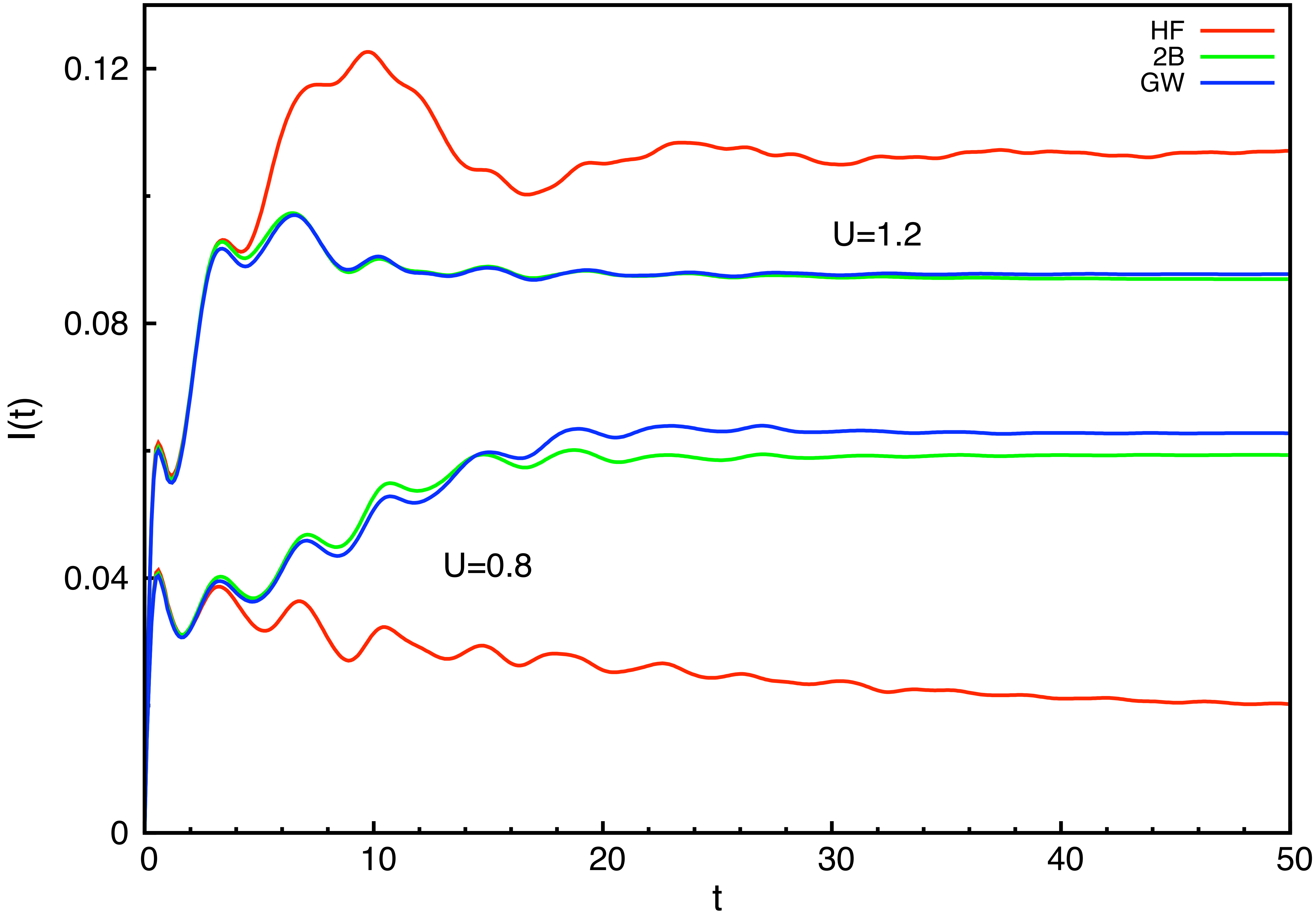


$$\mathcal{G}^{<}_{\mathrm{CC},HH}(t_1,t_2)$$



$$\mathcal{G}^{\rceil}_{\mathrm{CC},HH}(t,\tau)$$

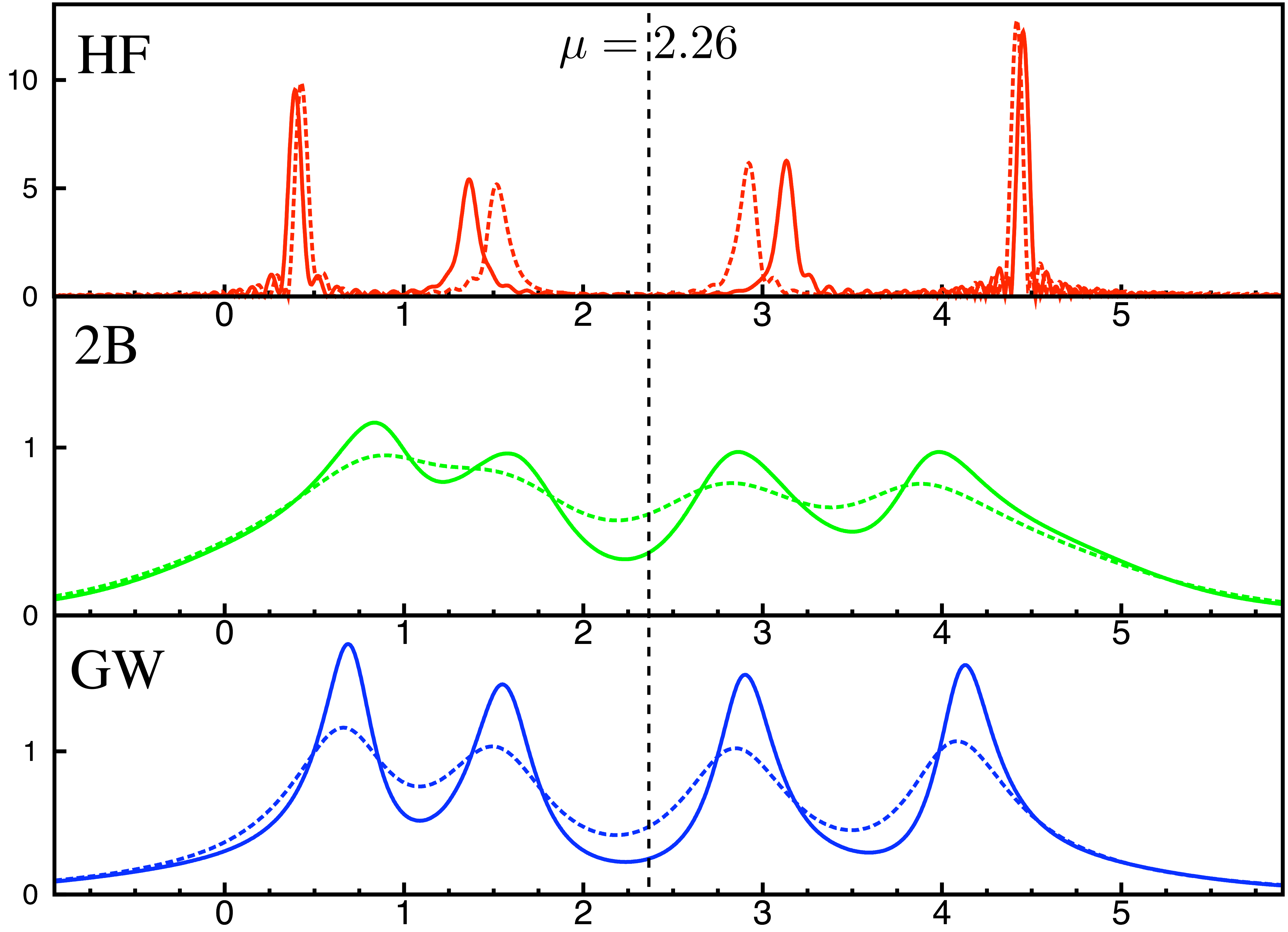
The transient currents



The spectral functions

Steady state regime

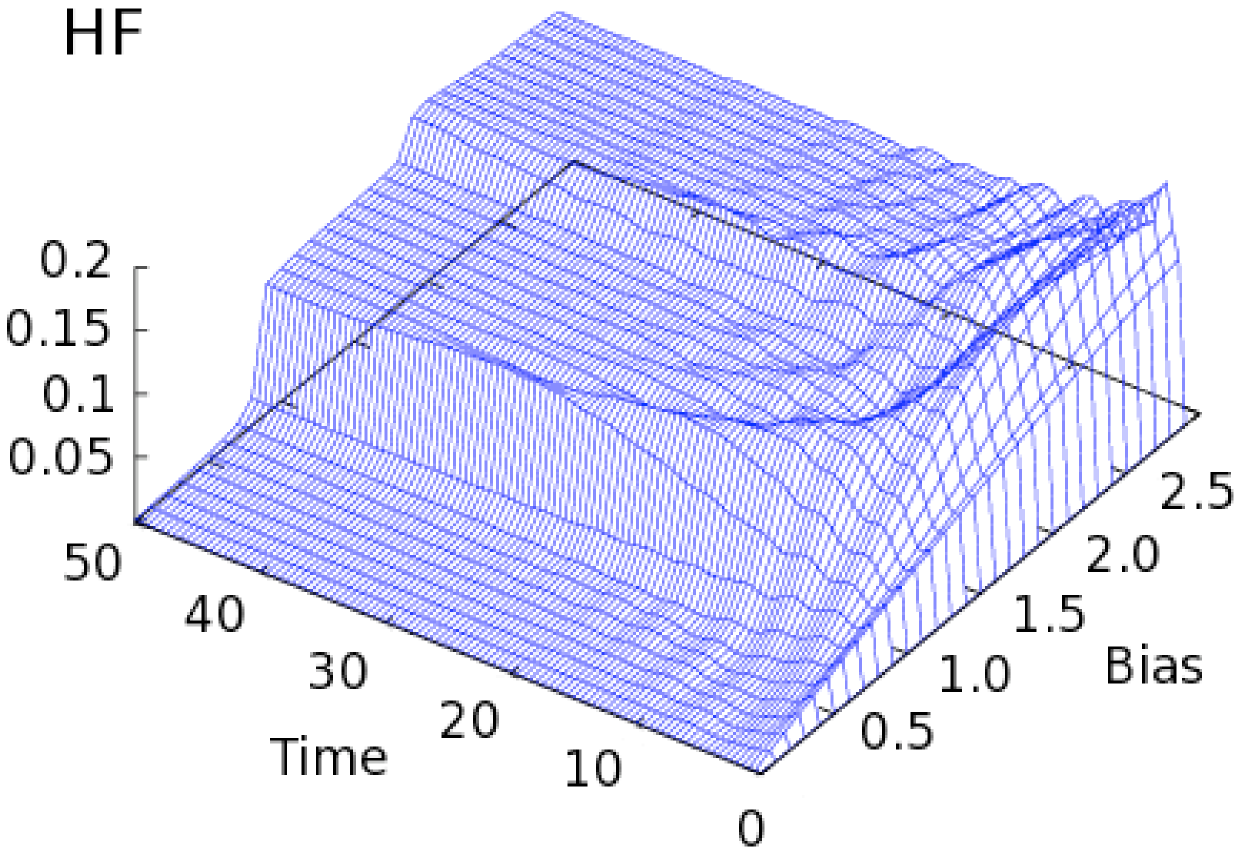
U=0.8 (solid line) U=1.2 (dashed line)

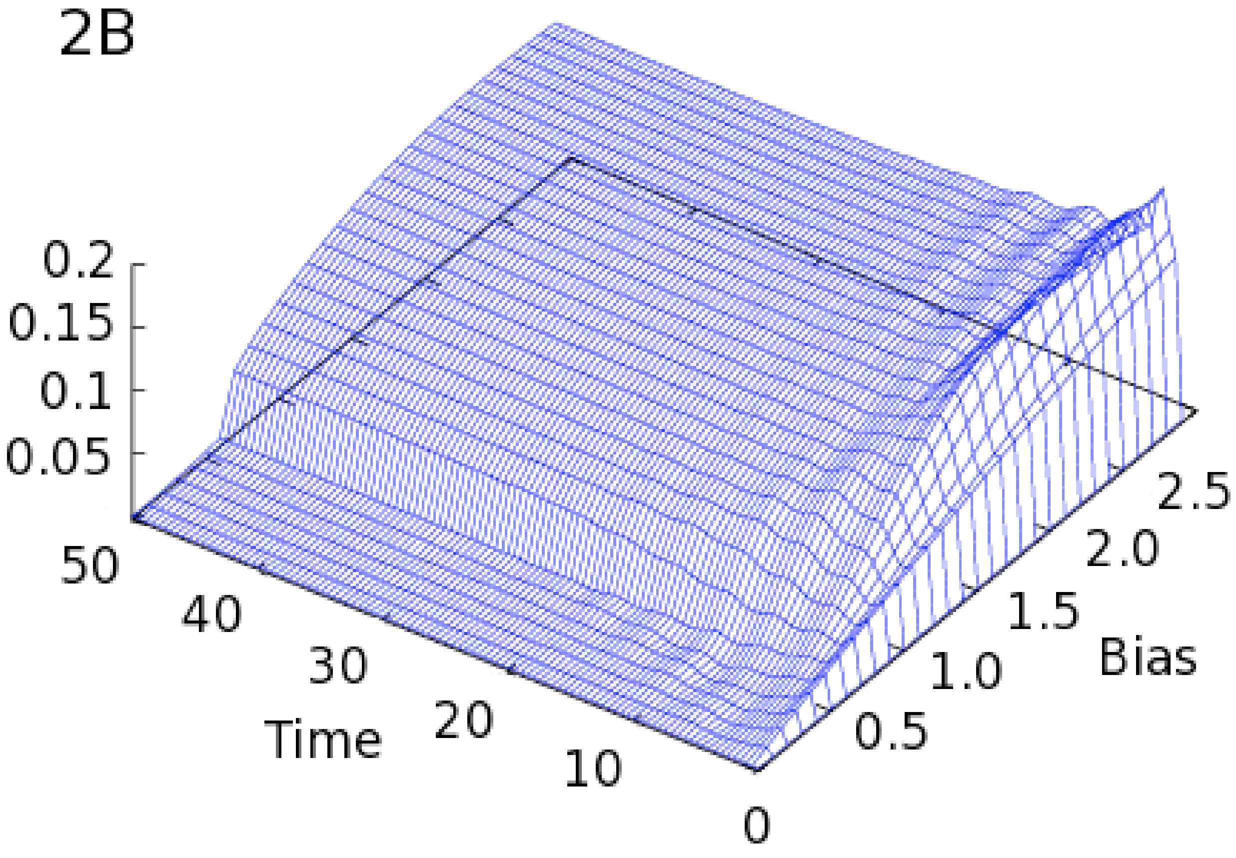


Time-dependent buildup of the I-V curves

Hartree-Fock

second Born

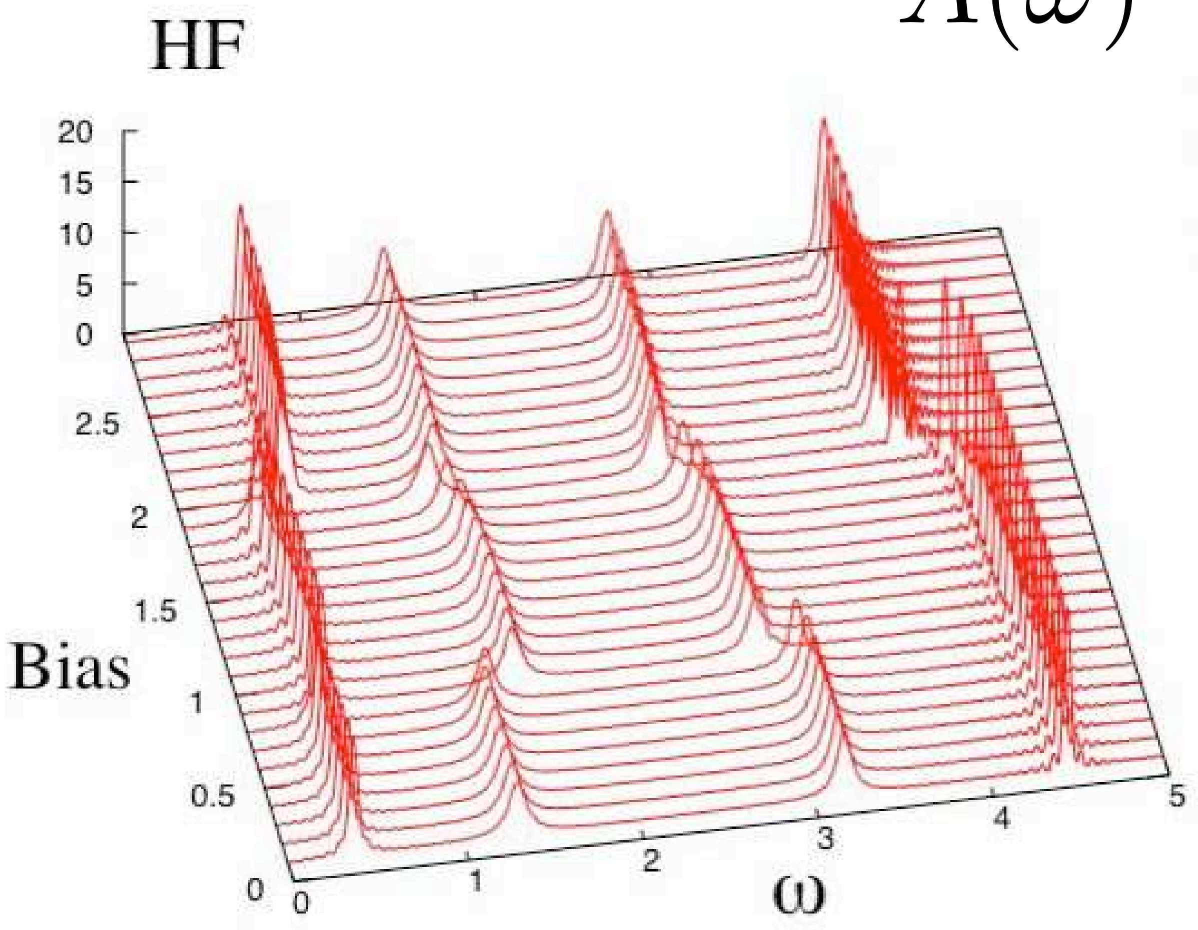


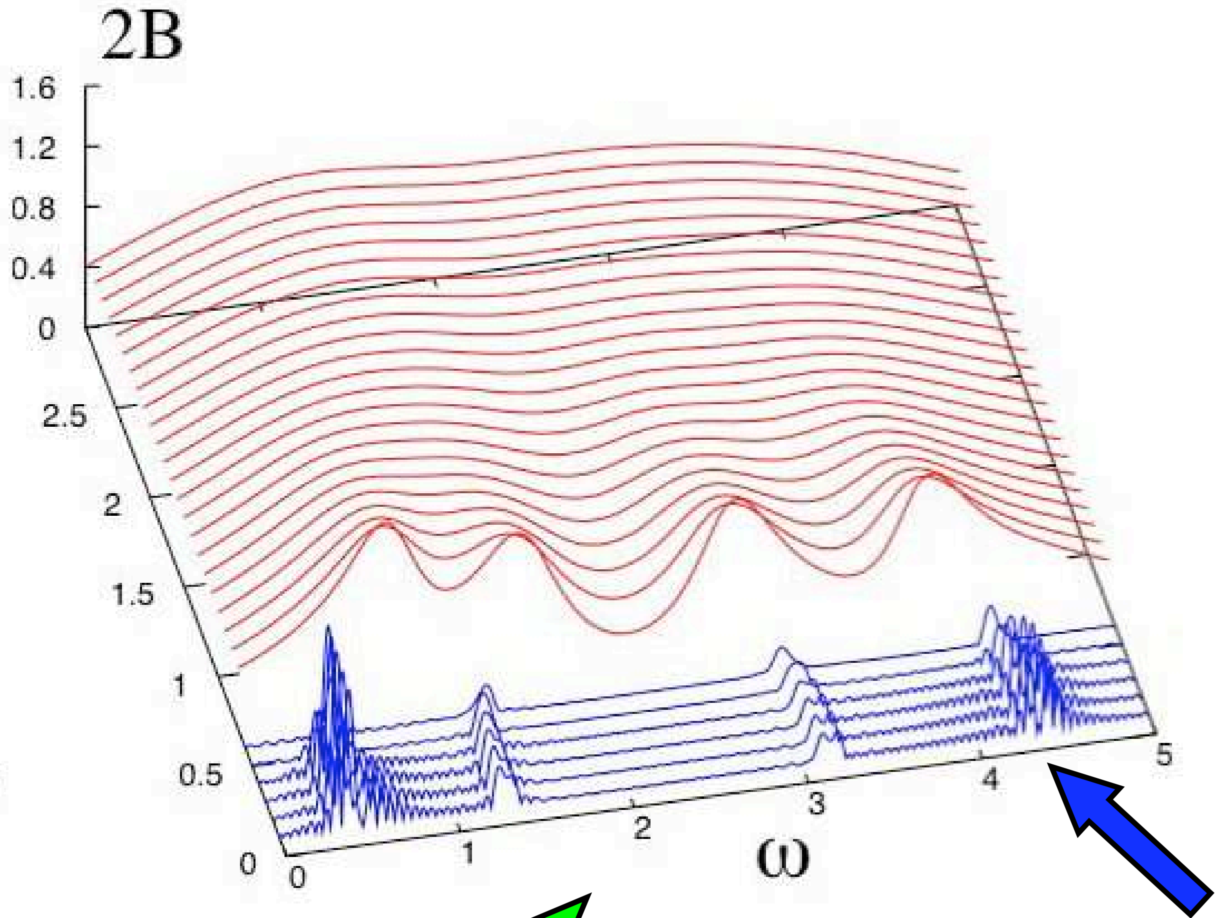


electron correlations beyond mean-field wash out I-V features

Bias dependence of the spectral functions

$A(\omega)$



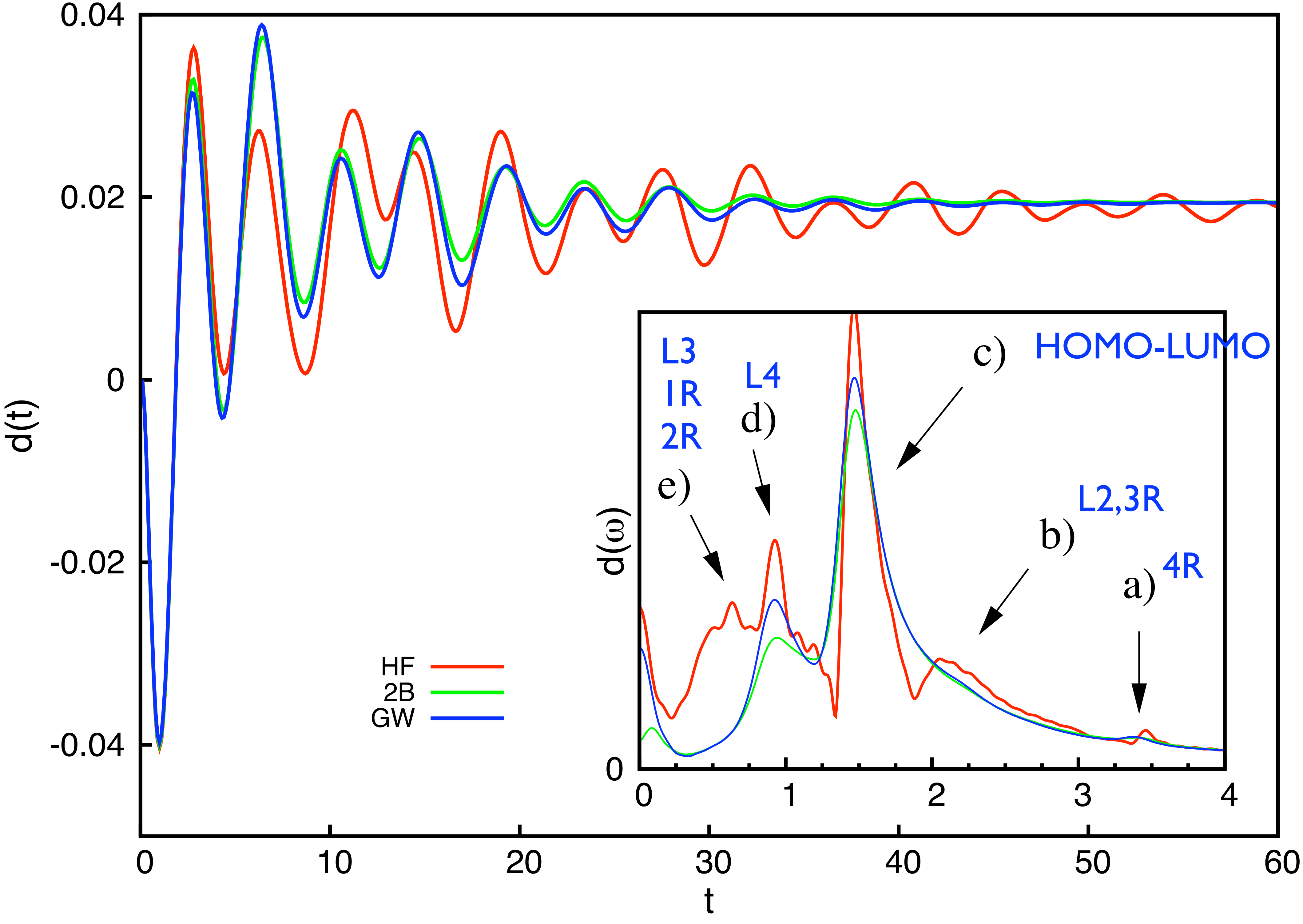


$A(\omega)/30$

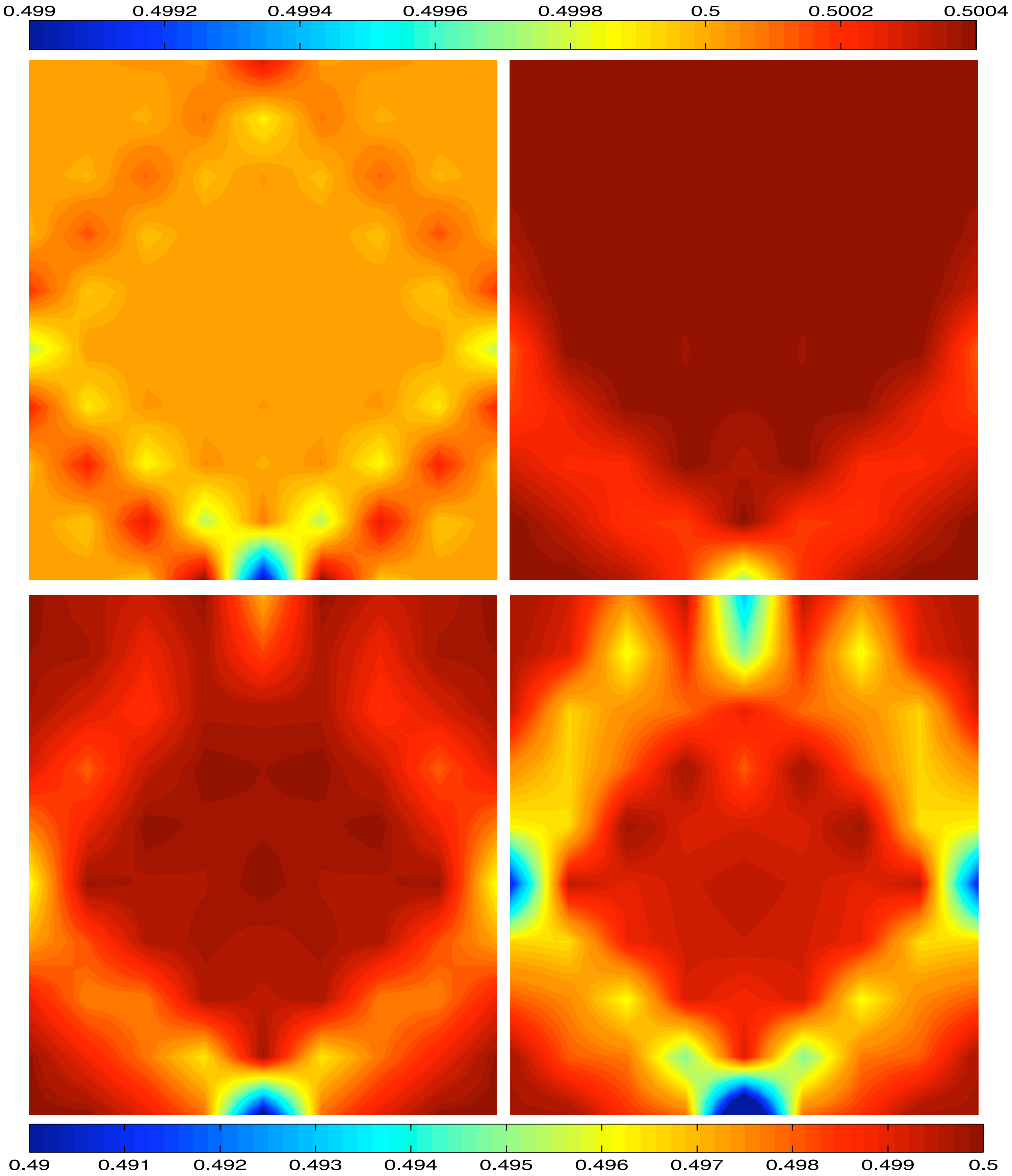
Gap closing

The time-dependent dipole moment

U=1.2

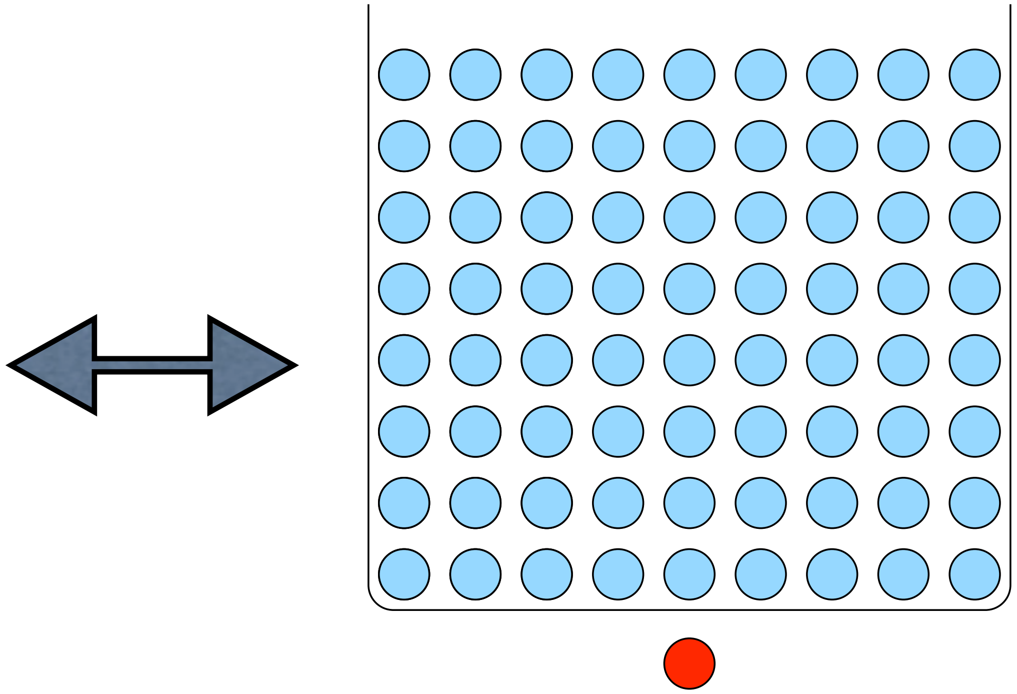


Time-dependent lead densities and Friedel oscillations



Charge flowing out of the left lead

(t=0, 1.7, 3.7, 10 au)



The density pattern can be understood from study of the density response function of the 2D tight binding lattice

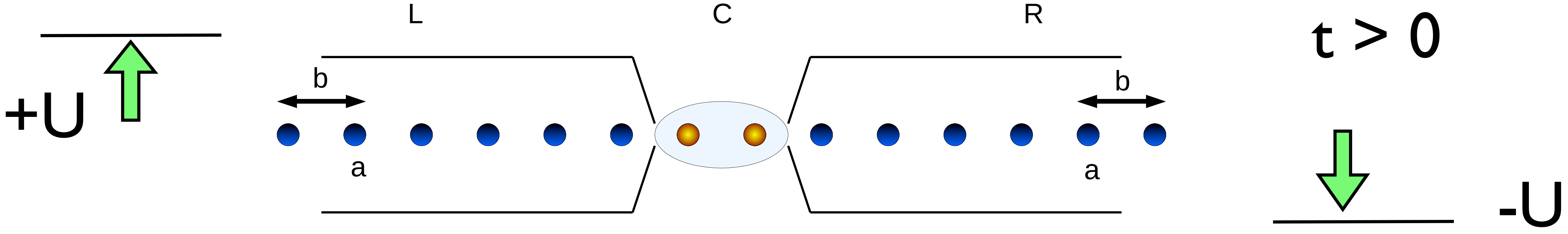
$$
\begin{aligned}
\chi(\mathbf{q},\omega) &= \int \frac{d\mathbf{k}}{(2\pi)^2} \frac{f(\epsilon_{\mathbf{k}}) - f(\epsilon_{\mathbf{k+q}})}{\omega - \epsilon_{\mathbf{k}} + \epsilon_{\mathbf{k+q}} + i\eta} \\
&= 2\int \frac{d\mathbf{k}}{(2\pi)^2} \frac{f(\epsilon_{\mathbf{k}})(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k+q}})}{(\omega + i\eta)^2 - (\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k+q}})^2}
\end{aligned}
$$

$$\epsilon_{\mathbf{k}} = 2T(\cos k_x + \cos k_y)$$

$\chi(\mathbf{q} = \alpha\mathbf{Q}, \omega = 0)$ with $\mathbf{Q} = (\pi, \pi)$

is discontinuous at $\alpha = 1$ leading to a cross-shaped density pattern

Model Hamiltonian



We use a correlated tight-binding Hamiltonian for the central region (2 sites) and an uncorrelated one for the leads

$$\hat{H}_C(t) = \sum_{i,j,\sigma} h_{ij}\, \hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + \sum_{ij\sigma\sigma'} v_{ij}\, \hat{c}^{\dagger}_{i\sigma}\hat{c}^{\dagger}_{j\sigma'}\hat{c}_{j\sigma'}\hat{c}_{i\sigma}$$

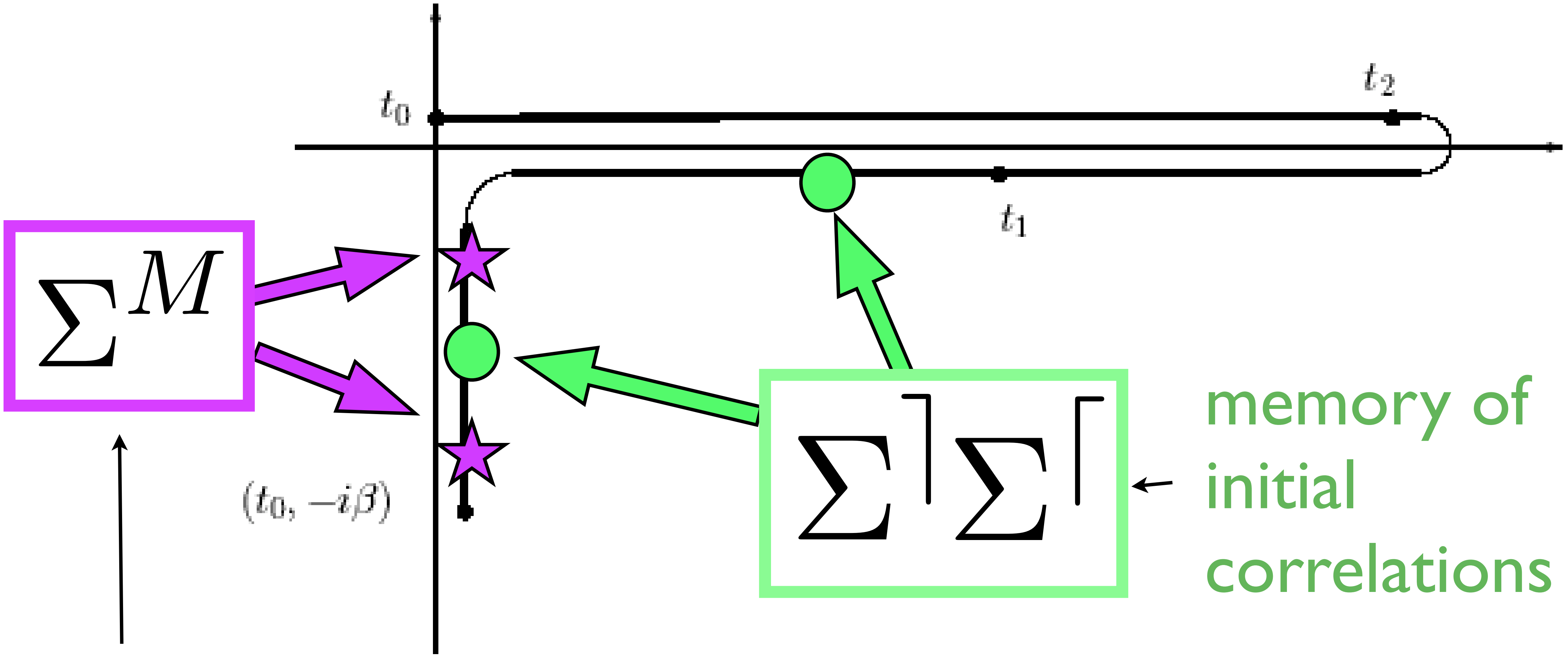
$$\hat{H}_\alpha(t) = \sum_{ij,\sigma}(t_{ij,\alpha} + \delta_{ij}U_\alpha(t))\hat{c}^{\dagger}_{i\alpha\sigma}\hat{c}_{j\alpha\sigma} \qquad U_\alpha(t) = \theta(t)\, U_\alpha$$

$$\hat{H}_{\text{coupling}} = \sum_{\alpha=L,R} H_{C\alpha} + \hat{H}_{\alpha C} = \sum_{\alpha=L,R}\sum_{jk,\sigma} V_{jk,\alpha}(\hat{c}^{\dagger}_{j\sigma}\hat{c}_{\alpha k\sigma} + \hat{c}^{\dagger}_{\alpha k\sigma}\hat{c}_{j\sigma})$$

(see also: Kristian Thygesen, PRL100, 166804 (2008))

Memory and initial correlations

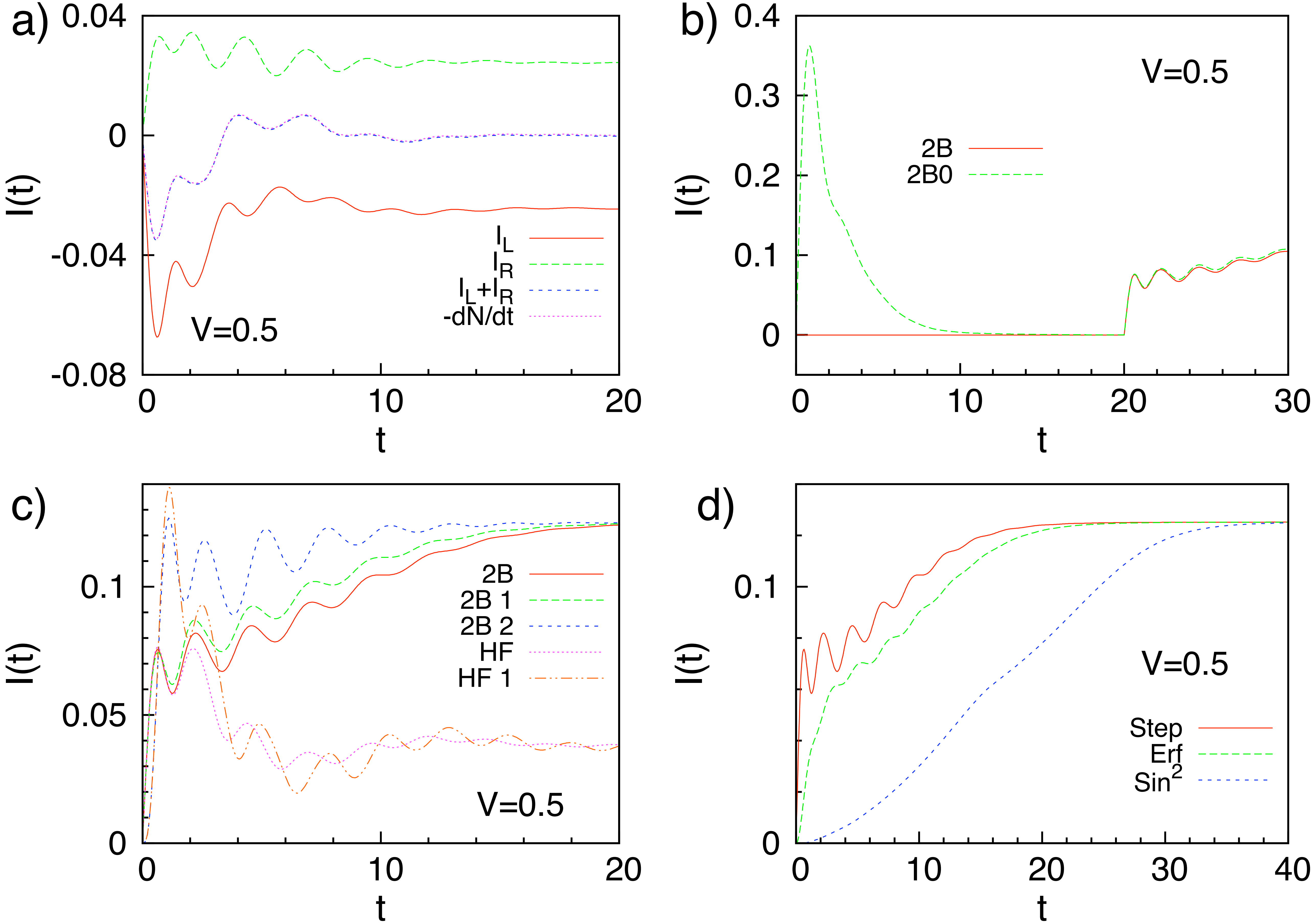
P.Myöhänen, A.Stan, G.Stefanucci, RvL
Europhys.Lett. 84, 67001 (2008)



memory of initial correlations

initial correlations

Memory and initial correlations



TDDFT : The challenge of a correlation functional with memory

The time-dependent xc-potential that gives the same density as that of the Kadanoff-Baym scheme, is given by (Sham-Schlüter equation)

$$\int dz' \, \boldsymbol{\mathcal{G}}_s(z,z')\boldsymbol{\mathcal{G}}(z',z)\mathbf{v}_{\mathrm{xc}}(z') = \int dz' dz'' \, \boldsymbol{\mathcal{G}}_s(z,z')\boldsymbol{\Sigma}[\boldsymbol{\mathcal{G}}](z',z'')\boldsymbol{\mathcal{G}}(z'',z)$$

This is not a closed equation unless we, for instance, make the substitution $\boldsymbol{\mathcal{G}} \longrightarrow \boldsymbol{\mathcal{G}}_s$

If this is done at Hartree-Fock level then we obtain the x-only TDOEP equations. The performance of this approach is likely to be close to TDHF.

What if the substitution is done at 2B or GW level?
Topic of future investigation......

Open problems

- Time-dependent quantum transport:
  Phonons (vibrons) + electronic interactions
  Two different time scales play a role, how to do this numerically?

- Finite systems:
  How to deal (at T=0) with degenerate initial states
  e.g. the ground state multiplet of the carbon atom ?

- Quantum transport:
  How do deal with lead interactions, avoiding reflections?

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MPI, Halle

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Kiel

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Thank you!

Conclusions

General conclusions

- An approach to the nonequilibrium quantum conduction problem is developed which is based on the solution of the Kadanoff-Baym equations for the nonequilibrium Green functions

- The scheme has build in conservations laws and effects of electron correlations can be explored by diagrammatic methods

- Macroscopic leads can be incorporated by means of embedding self-energies that are added on top of the self-energy terms that describe the electronic interactions

- Lead densities can be calculated from an inbedding self-energy

Conclusions for the 4 atom chain attached to 2D leads:

- Correlation effects beyond Hartree-Fock have a large influence on dynamics in quantum transport:

  a) At moderate bias the HOMO-LUMO gap closes while in HF it remains fairly constant

  b) The HOMO and LUMO resonances are rather sharp during the transient time and suddenly broaden when approaching the steady state. In HF they remain sharp.

  c) In the correlated case the transients are more damped and die out earlier

  d) Correlations beyond HF wash out features in I-V curves.

- For a 4-atom chain with long range interactions, screening effects are already considerable. The GW and 2B approximations for this case give very similar results

- All the oscillations in the TD dipole moment can be understood in terms of the level structure of the system. Transient spectroscopy?

Challenges:

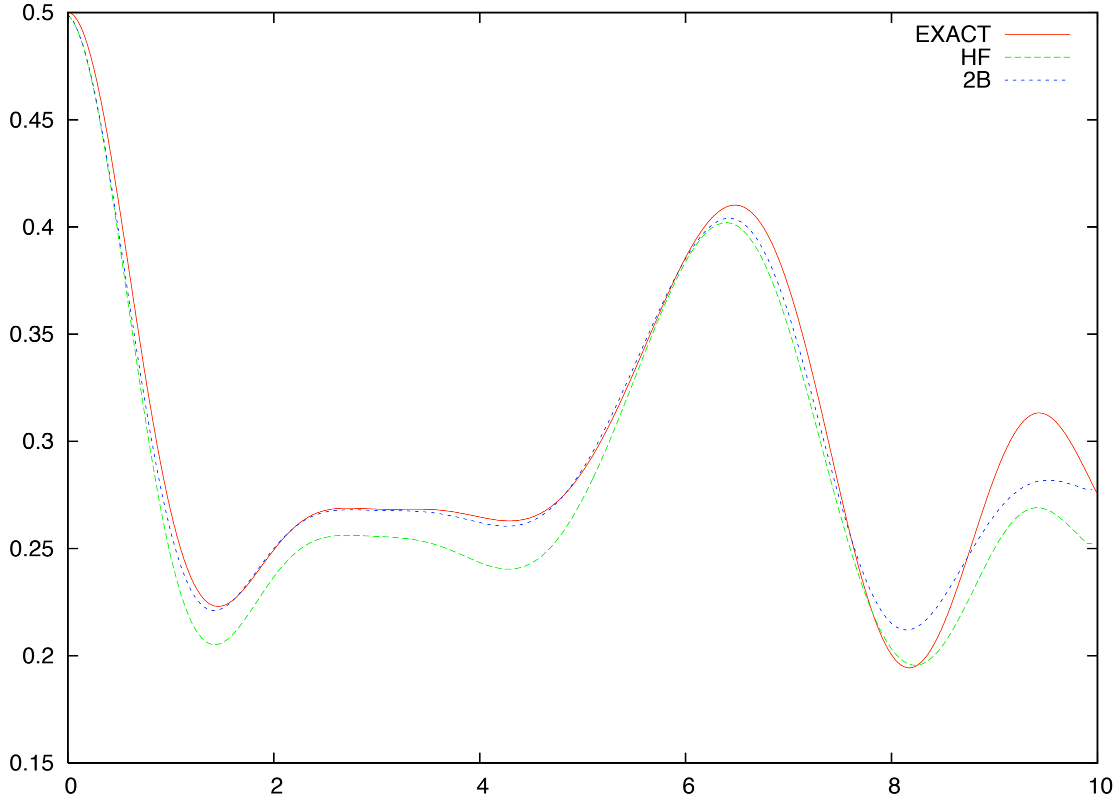
- Electron correlations + phonons:

  A problem of two time-scales

- Lead interactions (lead plasmons?)

-

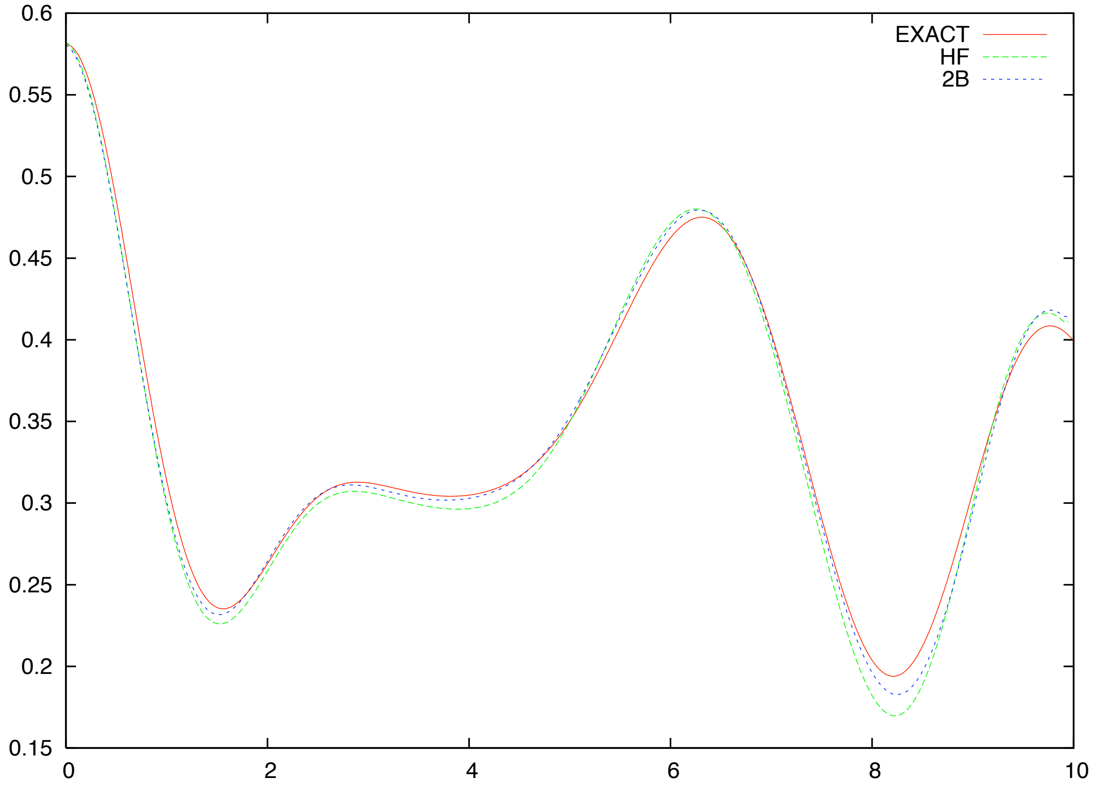
Comparison with exact solution TD Schrödinger equation



U=1, w=1

6 site Hubbard

(see also Marc Puig von Friesen, Claudio Verdozzi, Carl-Olof Almbladh, cond-mat 0905.2061)



6 site long-range Coulomb interaction