Speeding up GKBA calculations using initial correlations

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Kiel, March 2019

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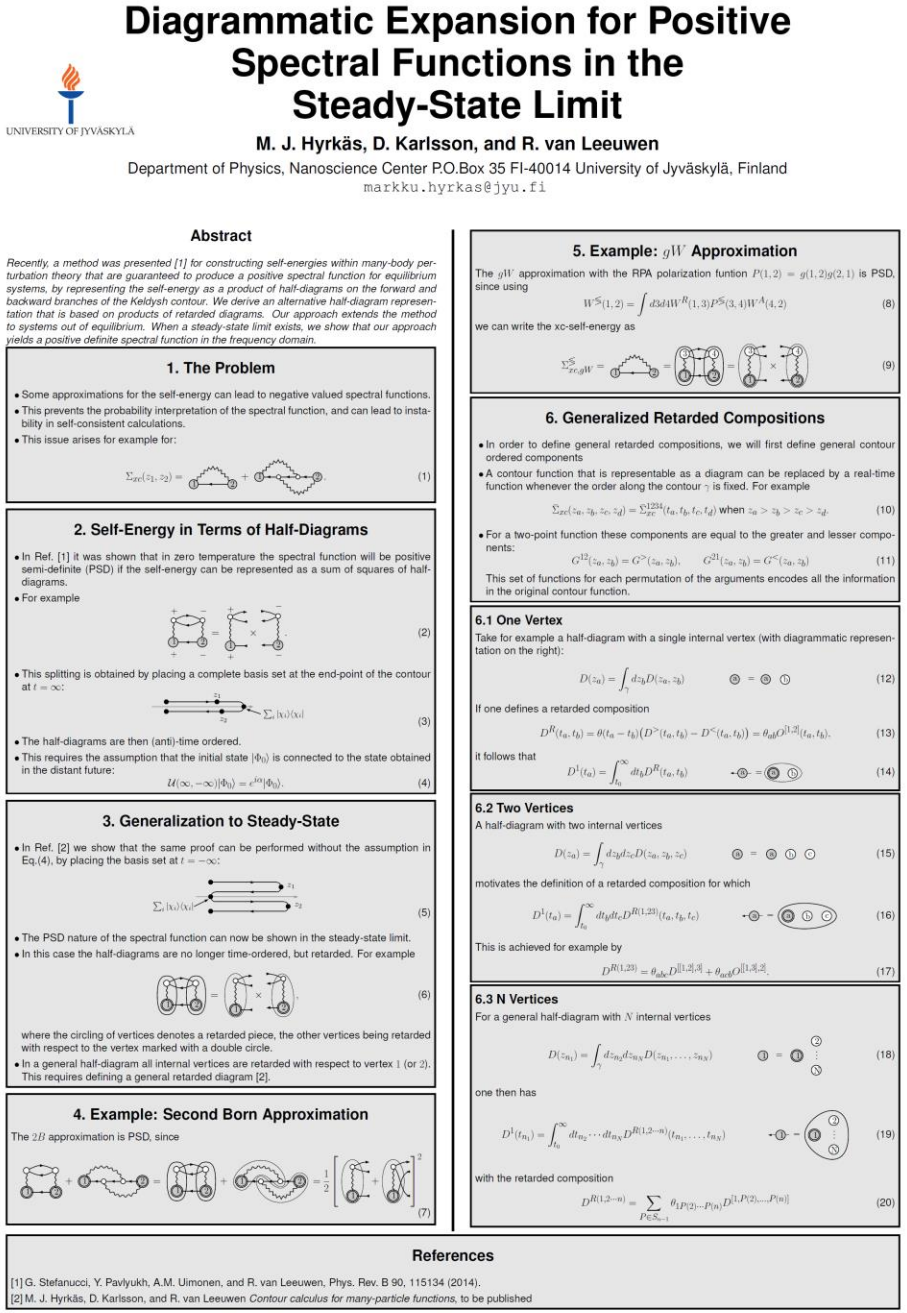
Positive steady-state spectral functions

- How to ensure positive spectral functions in steady-state when using diagrammatics?
- See Markku's poster!



Diagrammatic Expansion for Positive Spectral Functions in the Steady-State Limit

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Abstract

Recently, a method was presented [1] for constructing self-energies within many-body perturbation theory that are guaranteed to produce a positive spectral function for equilibrium systems, by representing the self-energy as a product of half-diagrams on the forward and backward branches of the Keldysh contour. We derive an alternative half-diagram representation that is based on products of retarded diagrams. Our approach extends the method to systems out of equilibrium. When a steady-state limit exists, we show that our approach yields a positive definite spectral function in the frequency domain.

1. The Problem

- Some approximations for the self-energy can lead to negative valued spectral functions.
- This prevents the probability interpretation of the spectral function, and can lead to instability in self-consistent calculations.
- This issue arises for example for:

$$\Sigma_{xc}(z_1,z_2) = \quad + \quad . \tag{1}$$

2. Self-Energy in Terms of Half-Diagrams

- In Ref. [1] it was shown that in zero temperature the spectral function will be positive semi-definite (PSD) if the self-energy can be represented as a sum of squares of half-diagrams.
- For example

$$= \quad \times \quad . \tag{2}$$

- This splitting is obtained by placing a complete basis set at the end-point of the contour at $t = \infty$:

$$\sum_i |\chi_i\rangle\langle\chi_i| \tag{3}$$

- The half-diagrams are then (anti)-time ordered.
- This requires the assumption that the initial state $|\Phi_0\rangle$ is connected to the state obtained in the distant future:

$$U(\infty,-\infty)|\Phi_0\rangle = e^{i\alpha}|\Phi_0\rangle. \tag{4}$$

3. Generalization to Steady-State

- In Ref. [2] we show that the same proof can be performed without the assumption in Eq.(4), by placing the basis set at $t = -\infty$:

$$\sum_i |\chi_i\rangle\langle\chi_i| \tag{5}$$

- The PSD nature of the spectral function can now be shown in the steady-state limit.
- In this case the half-diagrams are no longer time-ordered, but retarded. For example

$$= \quad \times \quad . \tag{6}$$

where the circling of vertices denotes a retarded piece, the other vertices being retarded with respect to the vertex marked with a double circle.

- In a general half-diagram all internal vertices are retarded with respect to vertex 1 (or 2). This requires defining a general retarded diagram [2].

4. Example: Second Born Approximation

The $2B$ approximation is PSD, since

$$+ \quad = \quad + \quad = \frac{1}{2}\left[\quad + \quad \right]^2 \tag{7}$$

5. Example: gW Approximation

The gW approximation with the RPA polarization funtion $P(1,2) = g(1,2)g(2,1)$ is PSD, since using

$$W^{\mathcal{S}}(1,2) = \int d3d4 W^R(1,3)P^{\mathcal{S}}(3,4)W^A(4,2) \tag{8}$$

we can write the xc-self-energy as

$$\Sigma^{\mathcal{S}}_{xc,gW} = \quad = \quad = \quad \times \quad . \tag{9}$$

6. Generalized Retarded Compositions

- In order to define general retarded compositions, we will first define general contour ordered components
- A contour function that is representable as a diagram can be replaced by a real-time function whenever the order along the contour γ is fixed. For example

$$\Sigma_{xc}(z_a,z_b,z_c,z_d) = \Sigma^{1234}_{xc}(t_a,t_b,t_c,t_d) \text{ when } z_a > z_b > z_c > z_d. \tag{10}$$

- For a two-point function these components are equal to the greater and lesser components:

$$G^{12}(z_a,z_b) = G^{>}(z_a,z_b), \qquad G^{21}(z_a,z_b) = G^{<}(z_a,z_b) \tag{11}$$

This set of functions for each permutation of the arguments encodes all the information in the original contour function.

6.1 One Vertex

Take for example a half-diagram with a single internal vertex (with diagrammatic representation on the right):

$$D(z_a) = \int_\gamma dz_b D(z_a,z_b) \qquad = \tag{12}$$

If one defines a retarded composition

$$D^R(t_a,t_b) = \theta(t_a-t_b)\big(D^>(t_a,t_b) - D^<(t_a,t_b)\big) = \theta_{ab}D^{[1,2]}(t_a,t_b), \tag{13}$$

it follows that

$$D^1(t_a) = \int_{t_0}^{\infty} dt_b D^R(t_a,t_b) \qquad = \tag{14}$$

6.2 Two Vertices

A half-diagram with two internal vertices

$$D(z_a) = \int_\gamma dz_b dz_c D(z_a,z_b,z_c) \qquad = \tag{15}$$

motivates the definition of a retarded composition for which

$$D^1(t_a) = \int_{t_0}^{\infty} dt_b dt_c D^{R(1,23)}(t_a,t_b,t_c) \qquad = \tag{16}$$

This is achieved for example by

$$D^{R(1,23)} = \theta_{ab\bar{c}}D^{[[1,2],3]} + \theta_{a\bar{b}\bar{c}}D^{[[1,3],2]}. \tag{17}$$

6.3 N Vertices

For a general half-diagram with N internal vertices

$$D(z_{a_1}) = \int_\gamma dz_{a_2}\cdots dz_{a_N} D(z_{a_1},\ldots,z_{a_N}) \qquad = \tag{18}$$

one then has

$$D^1(t_{a_1}) = \int_{t_0}^{\infty} dt_{a_2}\cdots dt_{a_N} D^{R(1,2\cdots n)}(t_{a_1},\ldots,t_{a_N}) \qquad = \tag{19}$$

with the retarded composition

$$D^{R(1,2\cdots n)} = \sum_{P\in S_{n-1}} \theta_{1P(2)\cdots P(n)} D^{[1,P(2),\ldots,P(n)]} \tag{20}$$

References

[1] G. Stefanucci, Y. Pavlyukh, A.M. Uimonen, and R. van Leeuwen, Phys. Rev. B 90, 115134 (2014).

[2] M. J. Hyrkäs, D. Karlsson, and R. van Leeuwen *Contour calculus for many-particle functions*, to be published

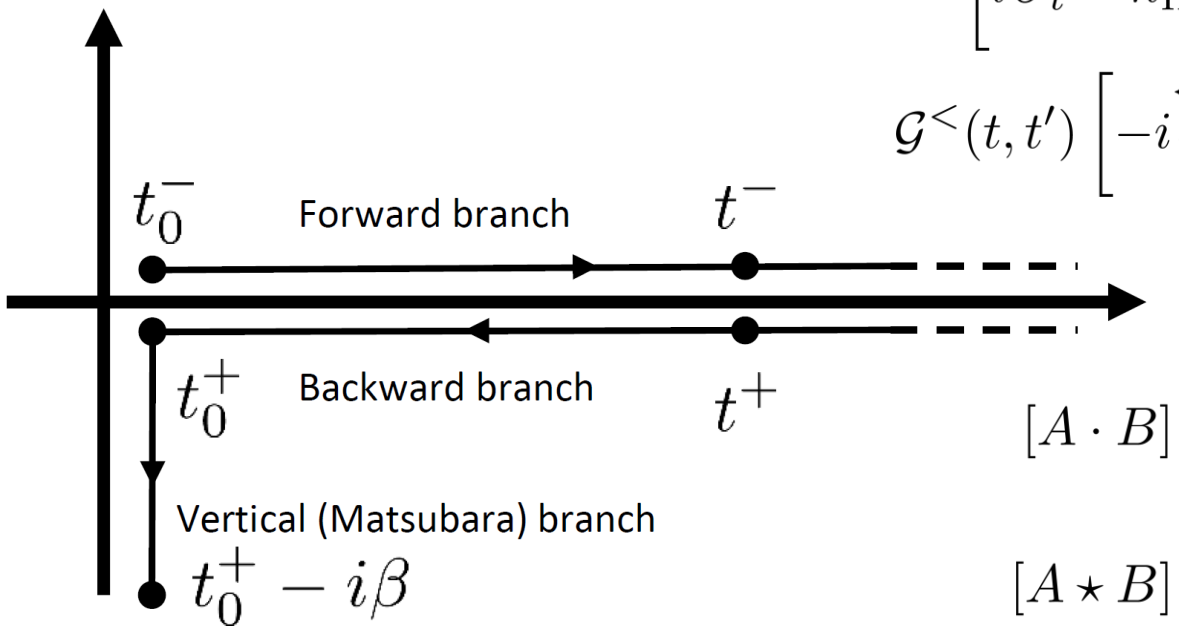
Outline

- The Kadanoff-Baym Equations
- The Generalized Kadanoff-Baym Ansatz (GKBA)
- Different strategies to include initial correlations within GKBA
  - Strategy 1: GKBA for imaginary times
  - Strategy 2: Write initial correlations in terms of real times
- Numerical example in an inhomogeneous system

Time-scaling problem of the Kadanoff-Baym Equations (KBE)

$$\left[i\vec{\partial}_t - h_{\mathrm{HF}}(t)\right]\mathcal{G}^<(t,t') = \left[\Sigma^< \cdot \mathcal{G}^A + \Sigma^R \cdot \mathcal{G}^< + \Sigma^\rceil \star \mathcal{G}^\lceil\right](t,t')$$

$$\mathcal{G}^<(t,t')\left[-i\overleftarrow{\partial}_{t'} - h_{\mathrm{HF}}(t')\right] = \left[\mathcal{G}^< \cdot \Sigma^A + \mathcal{G}^R \cdot \Sigma^< + \mathcal{G}^\rceil \star \Sigma^\lceil\right](t,t')$$



$$\varrho = e^{-\beta \hat{H}^M}$$

$$[A \cdot B](t,t') \equiv \int_0^\infty \mathrm{d}\bar{t}\, A(t,\bar{t})B(\bar{t},t') \text{ Real-time convolution}$$

$$[A \star B](t,t') \equiv -i\int_0^\beta \mathrm{d}\bar{\tau} A(t,\bar{\tau})B(\bar{\tau},t') \text{ Initial correlations}$$

$G(t,t')$ is two-time object, and involves real-time convolutions -> Scales like (nr of timesteps)^3: Difficult to treat numerically. Solution: Look at time diagonal!

From KBE to the equation of motion for ρ

Subtracting the two KBE equations and letting $t' \to t$ yields equation for density matrix $\rho(t) = -i\mathcal{G}^<(t,t)$:

$$\partial_t \rho(t) + i\left[h_{\mathrm{HF}}(t), \rho(t)\right] = -\left(\mathcal{I}(t) + \mathcal{I}^{\mathrm{ic}}(t) + \mathrm{H.c.}\right),$$

$$\mathcal{I}(t) = \int_0^t \mathrm{d}\bar{t}\left[\Sigma^>(t,\bar{t})\mathcal{G}^<(\bar{t},t) - \Sigma^<(t,\bar{t})\mathcal{G}^>(\bar{t},t)\right] \quad \textit{collision integral}$$

$$\mathcal{I}^{\mathrm{ic}}(t) = -i\int_0^\beta \mathrm{d}\bar{\tau}\,\Sigma^\rceil(t,\bar{\tau})\mathcal{G}^\lceil(\bar{\tau},t) \quad \textit{initial correlation integral}$$

- $\rho(t)$ is single-time object, and involves real-time convolutions -> Scales like (nr of timesteps)^2: one order of magnitude gained compared to KBE!
- Caveat: Not a closed equation for $\rho(t)$; Collision integral involves off-diagonal parts of G.
- Fix for real times: the GKBA

The GKBA

$$\partial_t \rho(t) + i\left[h_{\mathrm{HF}}(t), \rho(t)\right] = -\left(\mathcal{I}(t) + \mathcal{I}^{\mathrm{ic}}(t) + \mathrm{H.c.}\right),$$

$$\mathcal{I}(t) = \int_0^t \mathrm{d}\bar{t}\left[\Sigma^{>}(t,\bar{t})\mathcal{G}^{<}(\bar{t},t) - \Sigma^{<}(t,\bar{t})\mathcal{G}^{>}(\bar{t},t)\right] \quad \textit{collision integral}$$

In the collision integral, we make the approximation

$$\mathcal{G}^{<}(t,t') = -\left[\mathcal{G}^{R}(t,t')\rho(t') - \rho(t)\mathcal{G}^{A}(t,t')\right]$$
$$\mathcal{G}^{>}(t,t') = \left[\mathcal{G}^{R}(t,t')\bar{\rho}(t') - \bar{\rho}(t)\mathcal{G}^{A}(t,t')\right]$$

To close the equation, the retarded/advanced Green's function come from non-interacting system

$$\mathcal{G}^{R}(t,t') = -i\theta(t-t')\mathcal{T}\left\{e^{-i\int_{t'}^{t} h_{\mathrm{HF}}(\bar{t})d\bar{t}}\right\}$$

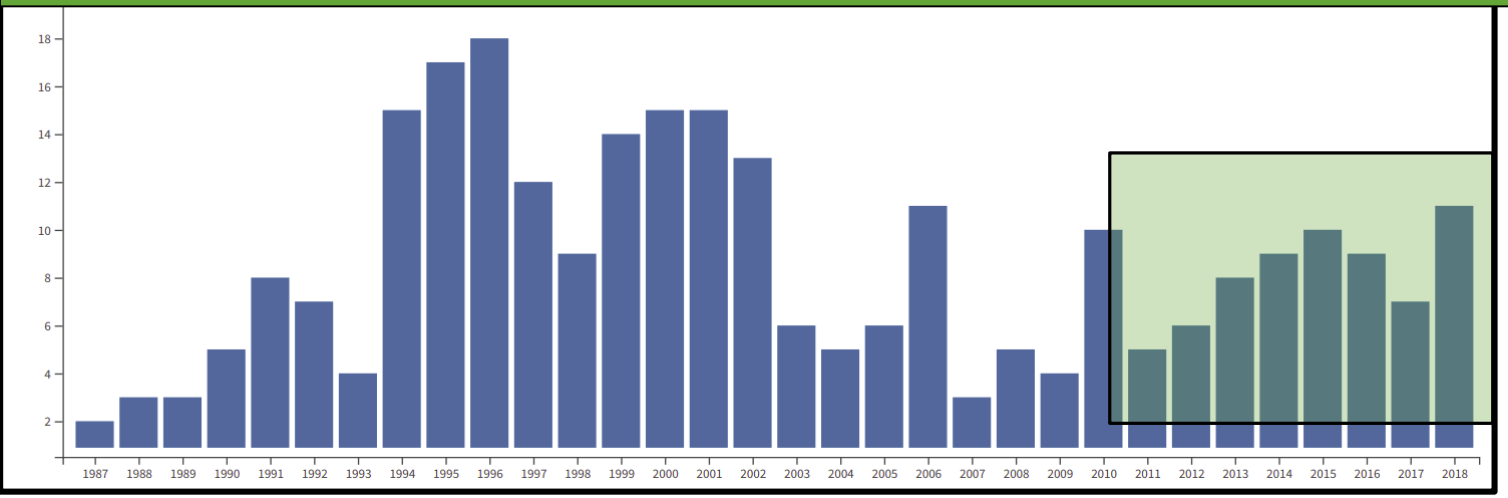
Useful manipulations within GKBA: 'Group property':

$$\mathcal{G}^{R}(t,t') = i\mathcal{G}^{R}(t,t_0)\mathcal{G}^{R}(t_0,t').$$

The GKBA

$$\mathcal{G}^{<}(t,t') = -\left[\mathcal{G}^{R}(t,t')\rho(t') - \rho(t)\mathcal{G}^{A}(t,t')\right]$$
$$\mathcal{G}^{>}(t,t') = \left[\mathcal{G}^{R}(t,t')\bar{\rho}(t') - \bar{\rho}(t)\mathcal{G}^{A}(t,t')\right]$$

P. Lipavský, V. Špička & B. Velický; *Generalized Kadanoff-Baym ansatz for deriving quantum transport equations.* PRB (1986)



The fast GKBA allows for studying:

Atoms

E. Perfetto, A.-M. Uimonen, R. van Leeuwen, and G. Stefanucci, PRA **92**, 033419 (2015).

Biologically relevant molecules

E. Perfetto, D. Sangalli, A. Marini, and G. Stefanucci, JCPL. **9**, 1353 (2018).

Organic compounds

G. Pal, Y. Pavlyukh,W. Hübner, and H. C. Schneider, EPJB **79**, 327 (2011).

E. V. Boström, A. Mikkelsen, C. Verdozzi, E. Perfetto, and G. Stefanucci, Nano Lett. **18**, 785 (2018).

Extended systems

D. Sangalli, S. Dal Conte, C. Manzoni, G. Cerullo, and A. Marini, PRB **93**, 195205 (2016).

Two-dimensional layered materials

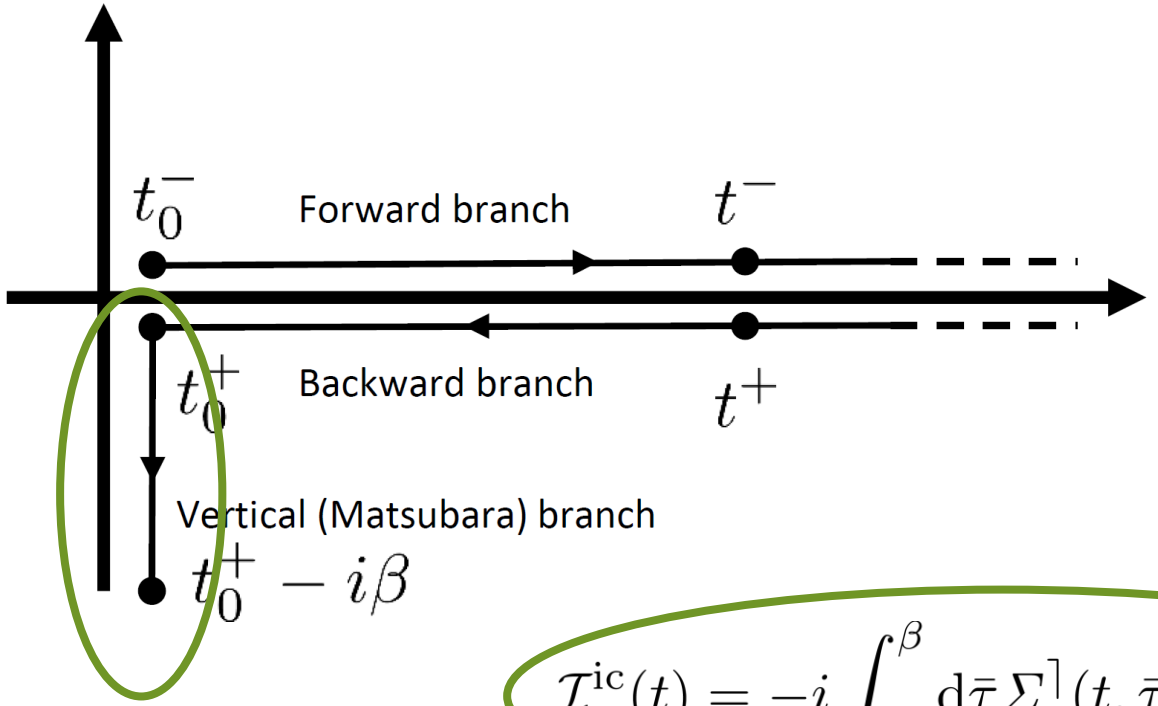
E. A. Pogna *et al* ACS Nano **10**, 1182 (2016).

A. Molina-Sánchez, D. Sangalli, L. Wirtz, and A. Marini, Nano Lett. **17**, 4549 (2017).

Doublon formation by ion impact

K. Balzer,M.R. Rasmussen, N. Schlünzen, J.-P. Joost, & M. Bonitz PRL **121**, 267602 (2018).

The issue with the GKBA



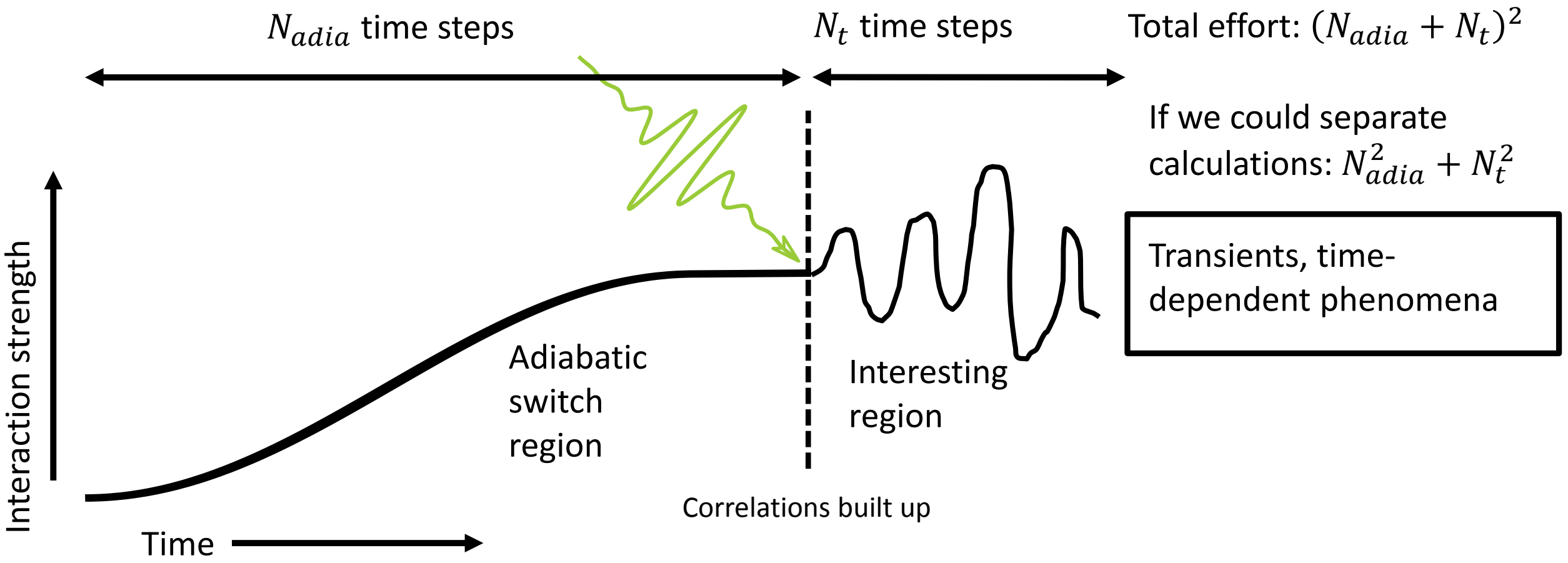
$$\mathcal{G}^<(t,t') = -\left[\mathcal{G}^R(t,t')\rho(t') - \rho(t)\mathcal{G}^A(t,t')\right],$$
$$\mathcal{G}^>(t,t') = \left[\mathcal{G}^R(t,t')\bar{\rho}(t') - \bar{\rho}(t)\mathcal{G}^A(t,t')\right],$$

Issue: GKBA Ansatz for real times only!
Initial correlations concerns imaginary-time.
No GKBA exists for imaginary times!

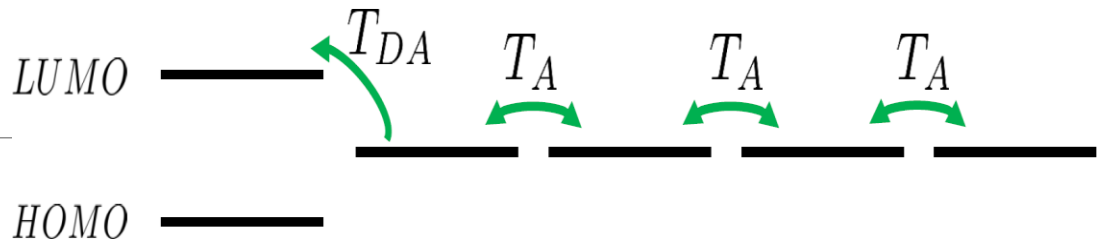
$$\mathcal{I}^{\rm ic}(t) = -i\int_0^\beta \mathrm{d}\bar{\tau}\,\Sigma^\rceil(t,\bar{\tau})\mathcal{G}^\lceil(\bar{\tau},t).$$

Common solution:
Start from non-interacting state!
$\Sigma^\rceil(t,\bar{\tau}) = 0$

Adiabatic switching technique



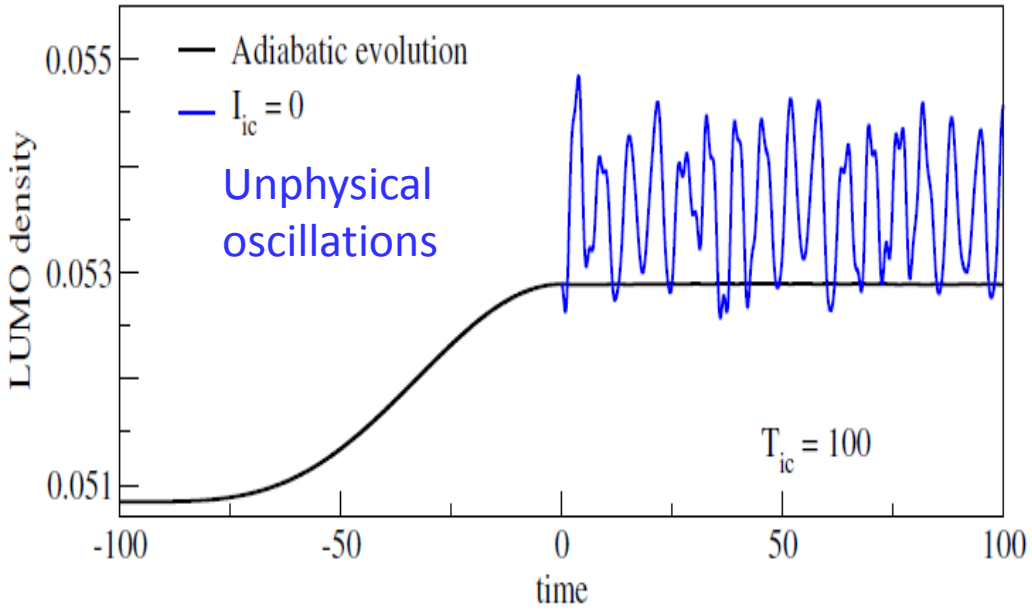
What happens if we anyway start with a correlated state?



Calculation 1: Adiabatic switch-on from T=-100

Calculation 2: Save density matrix ρ_{eq} from adiabatic switch at time t=0, then start another calculation from t=0 with $\rho(t=0) = \rho_{eq}$. IC term neglected.

$$\partial_t \rho(t) + i\left[h_{\mathrm{HF}}(t), \rho(t)\right] = -\left(\mathcal{I}(t) + \mathcal{I}^{\mathrm{ic}}(t) + \mathrm{H.c.}\right)$$



What went wrong? Stationarity condition:

$$\mathcal{I}^{\mathrm{ic}}(0) = \mathcal{I}(t) + \mathcal{I}^{\mathrm{ic}}(t)$$

Intricate cancellation from initial correlation term and collision integral

Specific goals

- Stationarity without external fields
- Obtain expression for initial correlations independent on adiabatic switching
- Expressions valid without assuming losses, or homogenous systems
- Basis-independent expressions
- Separate the time propagation from the obtaining of the initial state
- Efficient implementation in existing GKBA codes

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$$\mathcal{I}^{\rm ic}(t) = -i \int_0^\beta \mathrm{d}\bar{\tau}\, \Sigma^{\rceil}(t,\bar{\tau})\mathcal{G}^{\lceil}(\bar{\tau},t)$$

Initial correlations within GKBA: The **lesser** function $\mathcal{G}^<(t,t')$

Strategy 1: Make 'educated guess' to obtain GKBA for mixed Green's functions!
Go back to Dyson equation for the full contour:

$$\mathcal{G}(z,z') = \mathcal{G}_0(z,z') + \int_\gamma \mathrm{d}\bar{z}\mathrm{d}\bar{z}'\, \mathcal{G}(z,\bar{z})\Sigma_c(\bar{z},\bar{z}')\mathcal{G}_0(\bar{z}',z')$$

Use of Langreth rules yields, for the lesser function

$$\hat{\mathcal{G}}^<(t,t') = \hat{\mathcal{G}}^{\mathrm{R}}(t,t_0)\hat{\mathcal{G}}^<(t_0,t_0)\hat{\mathcal{G}}^{\mathrm{A}}(t_0,t') + \left[\hat{\mathcal{G}}^{\mathrm{R}}\cdot\left(\hat{\Sigma}_{\mathrm{c}}^{\rceil} + \hat{\Sigma}_{\mathrm{c}}^{\rceil}\star\mathcal{G}^{\mathrm{M}}\star\hat{\Sigma}_{\mathrm{c}}^{\lceil}\right)\cdot\hat{\mathcal{G}}^{\mathrm{A}}\right]$$
$$+ \mathrm{i}\hat{\mathcal{G}}^{\mathrm{R}}(t,t_0)\left[\hat{\mathcal{G}}^{\mathrm{M}}\star\hat{\Sigma}_{\mathrm{c}}^{\rceil}\cdot\hat{\mathcal{G}}^{\mathrm{A}}\right](t_0,t') - \mathrm{i}\left[\hat{\mathcal{G}}^{\mathrm{R}}\cdot\hat{\Sigma}_{\mathrm{c}}^{\lceil}\star\hat{\mathcal{G}}^{\mathrm{M}}\right](t,t_0)\hat{\mathcal{G}}^{\mathrm{A}}(t_0,t')$$

For non-interacting systems, only the first term is non-zero, and can be written in GKBA form

$$\mathcal{G}^<(t,t') = -\left[\mathcal{G}^R(t,t')\rho(t') - \rho(t)\mathcal{G}^A(t,t')\right]$$

using $\rho(t') = \mathcal{G}^R(t',t_0)\rho(t_0)\mathcal{G}^A(t_0,t')$

Initial correlations within GKBA: The **right** function $\mathcal{G}^{\rceil}(t,\tau)$

Strategy 1: Make 'educated guess' to obtain GKBA for right Green's function!
Go back to Dyson equation for the full contour:

$$\mathcal{G}(z,z') = \mathcal{G}_0(z,z') + \int_\gamma \mathrm{d}\bar{z}\mathrm{d}\bar{z}'\, \mathcal{G}(z,\bar{z})\Sigma_c(\bar{z},\bar{z}')\mathcal{G}_0(\bar{z}',z')$$

Use of Langreth rules yields, for the right function

$$\mathcal{G}^{\rceil}(t,\tau) = i\mathcal{G}^R(t,t_0)\mathcal{G}^M(0,\tau) + \left[\mathcal{G}^R \cdot \Sigma_c^{\rceil} * \mathcal{G}^M\right](t,\tau)$$

For non-interacting systems, only first term is non-zero. GKBA for initial correlations?

Fails. All simple ways to do so gives problems (lack of stationarity in absense of external fields)

Strategy 2: Rewrite the initial correlations in terms of real times, and then apply usual GKBA

The initial correlation term in equilibrium

Inspiration: In equilibrium for t $\leq t_0 = 0$, from the exact KBE equations, $\mathcal{I}^{\rm ic}(t) = -i\int_0^\beta \mathrm{d}\bar{\tau}\Sigma^{\rceil}(t,\bar{\tau})\mathcal{G}^{\lceil}(\bar{\tau},t)$

G. Stefanucci, R. van Leeuwen, *Nonequilibrium Many-Body Theory of Quantum Systems* (2013)

$$\mathcal{I}^{\rm ic}(t) = \int_{-\infty}^{0} \mathrm{d}\bar{t}\left[\Sigma^{>}(t-\bar{t})\mathcal{G}^{<}(\bar{t}-t) - \Sigma^{<}(t-\bar{t})\mathcal{G}^{>}(\bar{t}-t)\right]$$

Proven via

$$\mathcal{G}^{\lceil}(\tau,t_2) = e^{\mu\tau}\mathcal{G}^{>}(-i\tau,t_2)$$
$$\mathcal{G}^{<}(\omega) = -e^{-\beta(\omega-\mu)}\mathcal{G}^{>}(\omega)$$

Equilibrium

Analytical continuation

Fluctuation-dissipation theorem

D. Karlsson, R. van Leeuwen, E. Perfetto, G. Stefanucci, PRB (2018)

$$\mathcal{I}^{\rm ic}(t) = \int_{-\infty}^{0} \mathrm{d}\bar{t}\left[\Sigma^{>}(t,\bar{t})\mathcal{G}^{<}(\bar{t},t) - \Sigma^{<}(t,\bar{t})\mathcal{G}^{>}(\bar{t},t)\right]$$

Proven via

$$\mathcal{G}^{\lceil}(\tau,t_2) = e^{\mu\tau}\mathcal{G}^{>}_{\rm aux}(-i\tau,t_2)$$
$$\mathcal{G}^{<}_{\rm aux}(\omega,t') = -e^{-\beta(\omega-\mu)}\mathcal{G}^{>}_{\rm aux}(\omega,t')$$

Out of equilibrium

Using ”half” of fluctuation-dissipation theorem

Initial correlations, explicit expressions

$$\mathcal{I}^{\rm ic}(t) = \int_{-\infty}^{0} {\rm d}\bar{t}\left[\Sigma^{>}(t,\bar{t})\mathcal{G}^{<}(\bar{t},t) - \Sigma^{<}(t,\bar{t})\mathcal{G}^{>}(\bar{t},t)\right]$$

Same shape as for adiabatic switching, but more general!
Valid for closed systems, finite temp etc.

Adiabatic switchings (or losses) + homogeneous systems: Restart technique exists. For a review, see: Bonitz, M., Balzer, K., Schlünzen, N., Rasmussen, M. R., & Joost, J.-P. PSSB (2019)

We can apply GKBA, with HF propagators or general propagators!
For general propagators, one can prove stationarity condition!

$$\mathcal{I}^{\rm ic}(0) = \mathcal{I}(t) + \mathcal{I}^{\rm ic}(t)$$

But, we still have to do time integrations.

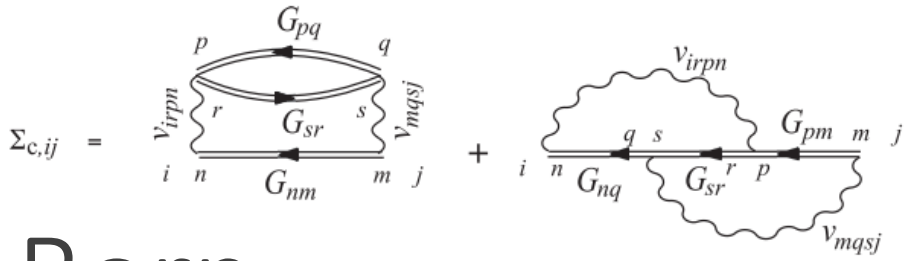
Use group property of HF propagators: $\mathcal{G}^R(t,\bar{t}) = i\mathcal{G}^R(t,0)\mathcal{G}^R(0,\bar{t})$

Hamiltonian constant for $\bar{t} \leq 0$ (equilibrium): $\mathcal{G}^R(0,\bar{t}) = -ie^{ih^{\rm eq}_{\rm HF}\bar{t}}$ Allows for analytical calculations

We obtained two numerically efficient cases within GKBA:
Case 1: 2nd Born. Case 2: GW_0, where W_0 taken from equilibrium calculations.

$$\Sigma^{<}_{ij}(t,\bar{t}) = \sum_{mnpqrs} v_{irpn}(t) w_{mqsj}(\bar{t}) \mathcal{G}^{<}_{nm}(t,\bar{t}) \mathcal{G}^{<}_{pq}(t,\bar{t}) \mathcal{G}^{>}_{sr}(\bar{t},t).$$

$$w_{imnj}(t) \equiv 2v_{imnj}(t) - v_{imjn}(t)$$



Initial correlations for 2nd Born

- Obtain equilibrium density matrix ρ^{eq}
- Calculate $h^{\mathrm{eq}}_{\mathrm{HF}} = h_{\mathrm{HF}}[\rho^{\mathrm{eq}}]$.
- Diagonalize $h^{\mathrm{eq}}_{\mathrm{HF}}$ to obtain eigenvalues ϵ_i

$$\tilde{\mathcal{I}}^{\mathrm{ic}}_{ik}(t) = i \sum_{npr} \frac{\tilde{v}_{irpn}(t)\tilde{w}_{nprk}}{\epsilon_r + \epsilon_k - \epsilon_n - \epsilon_p}$$

The tensor $\tilde{w}$ (Calculated once!) is

$$\tilde{w} \equiv \sum w \left(\bar{\rho}^{\mathrm{eq}} \bar{\rho}^{\mathrm{eq}} \rho^{\mathrm{eq}} \rho^{\mathrm{eq}} - \rho^{\mathrm{eq}} \rho^{\mathrm{eq}} \bar{\rho}^{\mathrm{eq}} \bar{\rho}^{\mathrm{eq}} \right).$$

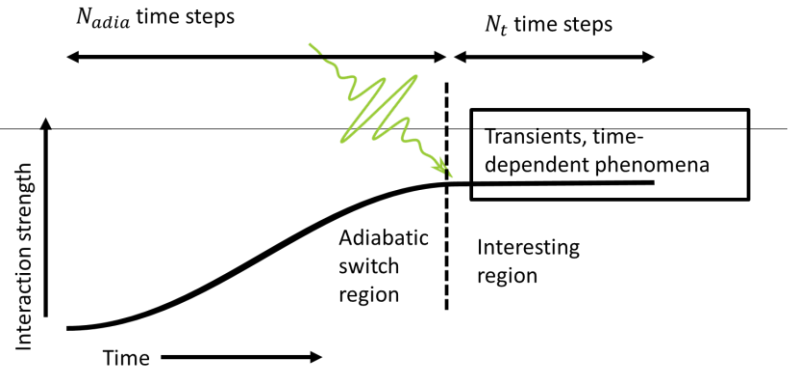
All time-dependence isolated to tensor $\tilde{v}(t)$

$$\tilde{v}(t) \equiv \sum v(t)\, \mathcal{G}^{R}(t,0) \mathcal{G}^{R}(t,0) \mathcal{G}^{A}(0,t).$$

Explicitly: $\mathcal{I}^{\mathrm{ic}} = \mathcal{I}^{\mathrm{ic}}[\rho]$. No time integration! Scales like (basis size)^5, just like original 2nd Born approximation!

How to obtain initial ρ^{eq}?

- The adiabatic switching technique
  - Works well, speeds up calculations compared to usual GKBA
  - Not always possible (finite temperature, phase transitions etc.)



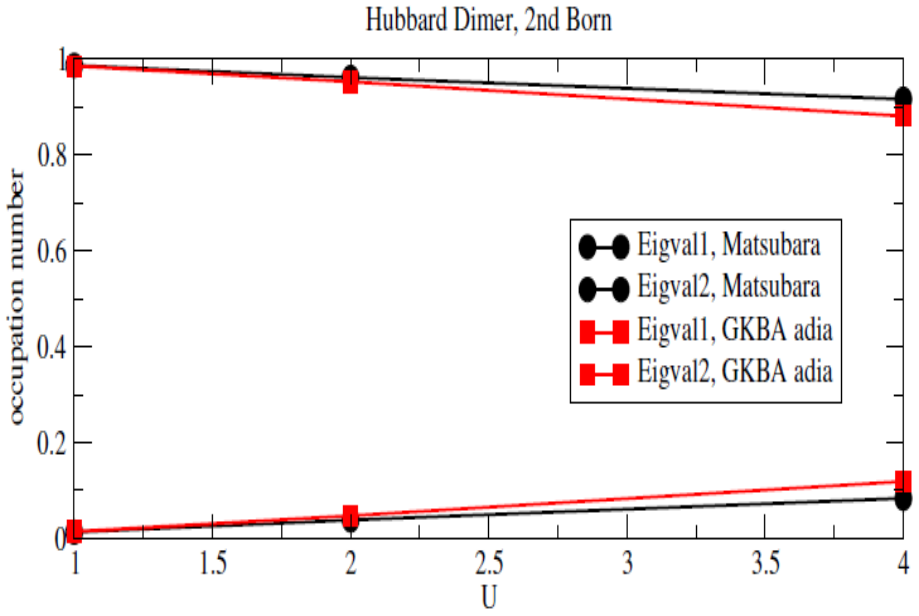
- Use equation of motion for ρ at t=0

$$-i\left[h^{\mathrm{eq}}_{\mathrm{HF}}, \rho^{\mathrm{eq}}\right] = \mathcal{I}^{\mathrm{ic}}(0) + \mathrm{H.c.}.$$

  - Supplement with reasonable occupations.

$$\rho^{\mathrm{eq},(i+1)}_{nn} = f_n$$

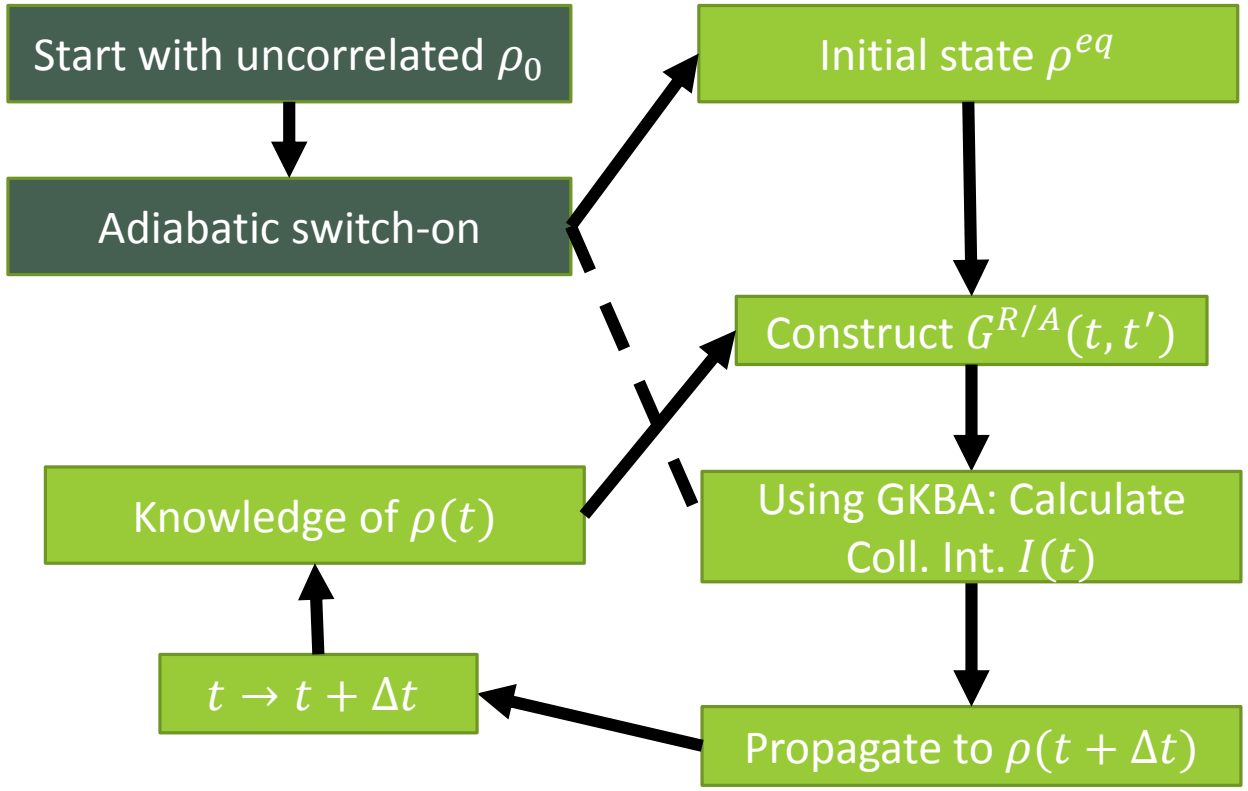
For example, from Matsubara calculation. Can be solved iteratively.



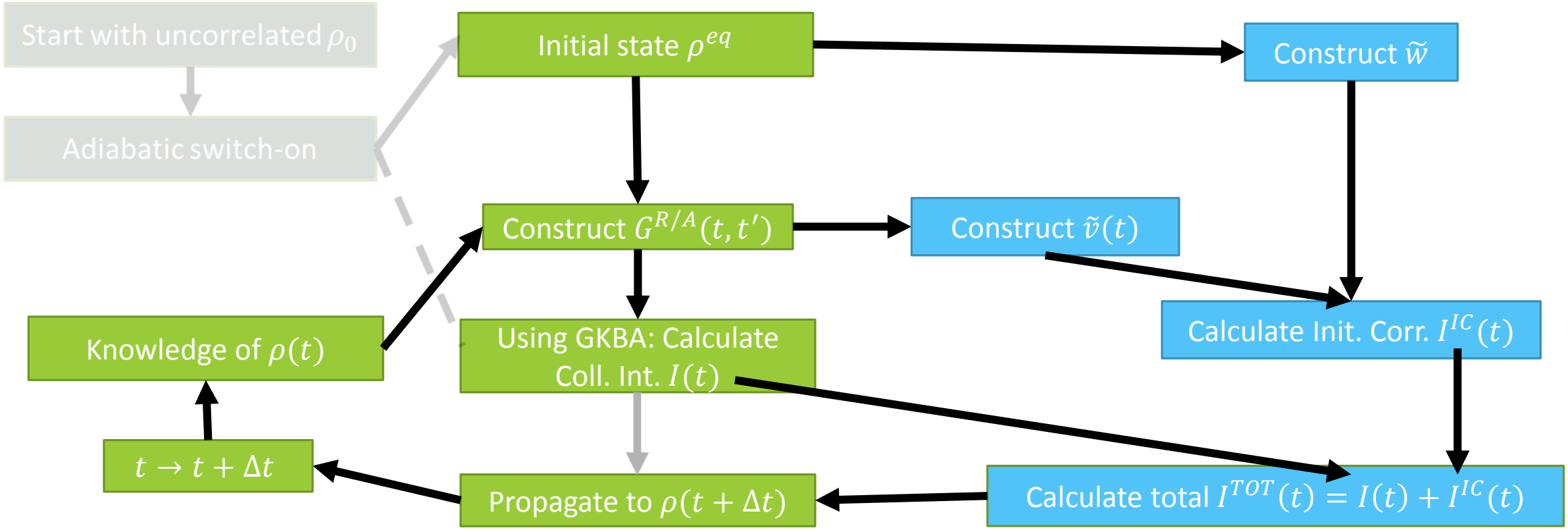
- Other methods of obtaining ρ,

  see Hopjan, M. & Verdozzi, C. Eur. Phys. J. Spec. Top. (2019).

Flowchart of GKBA calculation with adiabatic switch-on



Flowchart of GKBA calculation with initial correlations

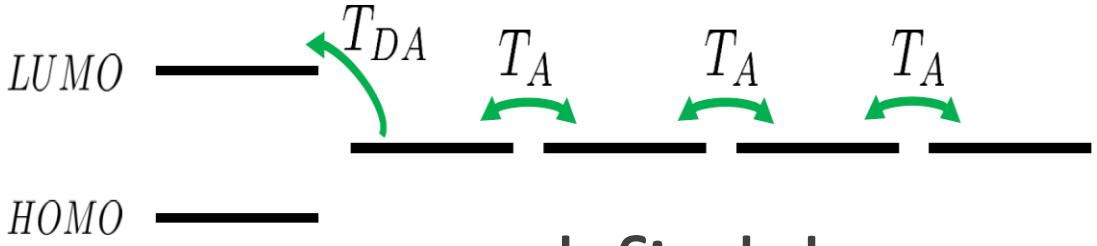


Initial correlations developed in the CHEERS code

E. Perfetto & G. Stefanucci, J. Phys.: Condens. Matter (2018)

Specific goals

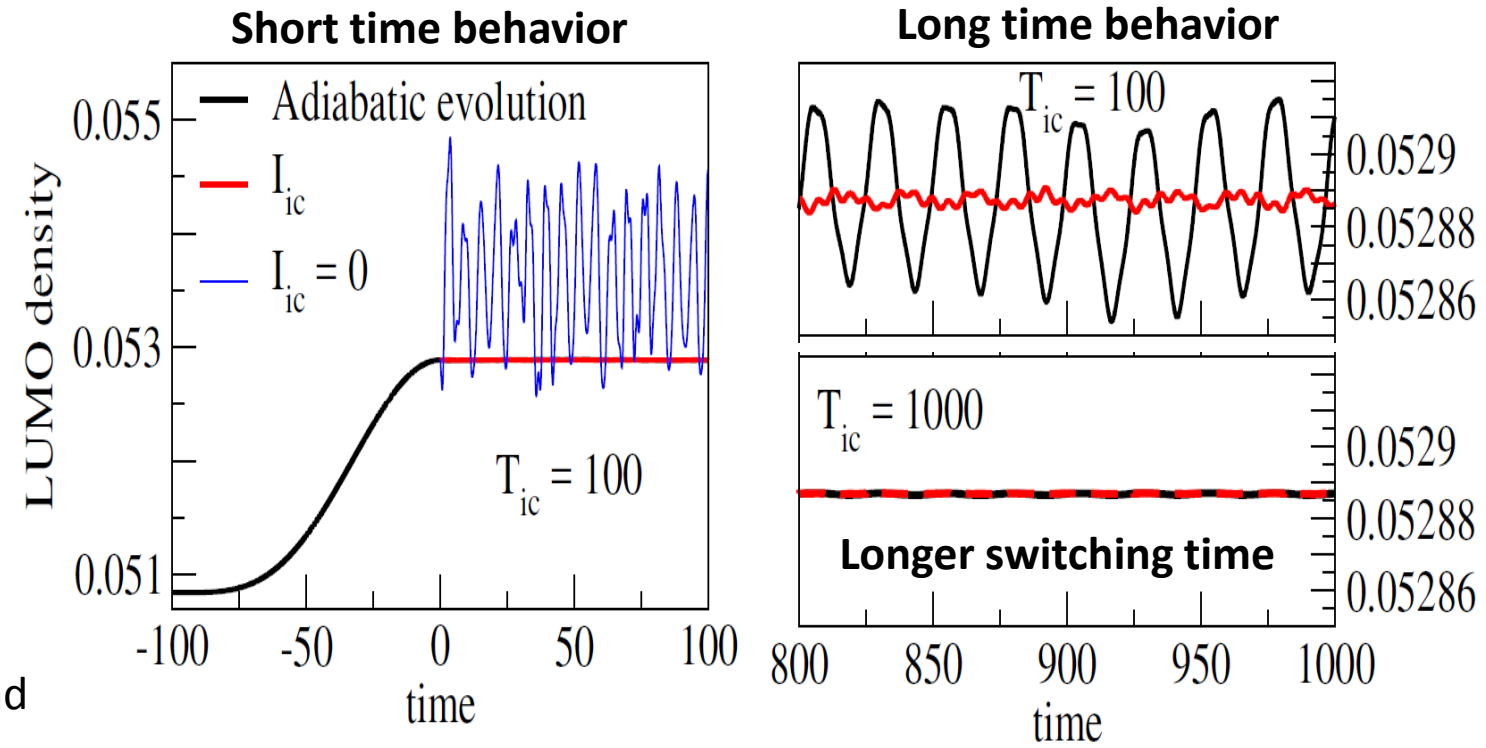
- ✓ Obtain expression for initial correlations independent on adiabatic switching
- ✓ Expressions valid without assuming losses, or homogenous systems
- ✓ Basis-independent expressions
- ✓ Separate the time propagation from the obtaining of the initial state
- ✓ Efficient implementation in existing GKBA codes



Initial correlations without external fields

Procedure: Generate ρ_{eq} with adiabatic switching with time T_{ic}. Then restart calculation from $t > 0$ with $I^{ic}[\rho, \rho_{eq}]$.

- Neglecting I^{ic} introduces large error
- Procedure is stable, even if ρ_{eq} not perfectly generated
- Procedure numerically efficient: I^{ic} and $I^{ic} = 0$ has same cost



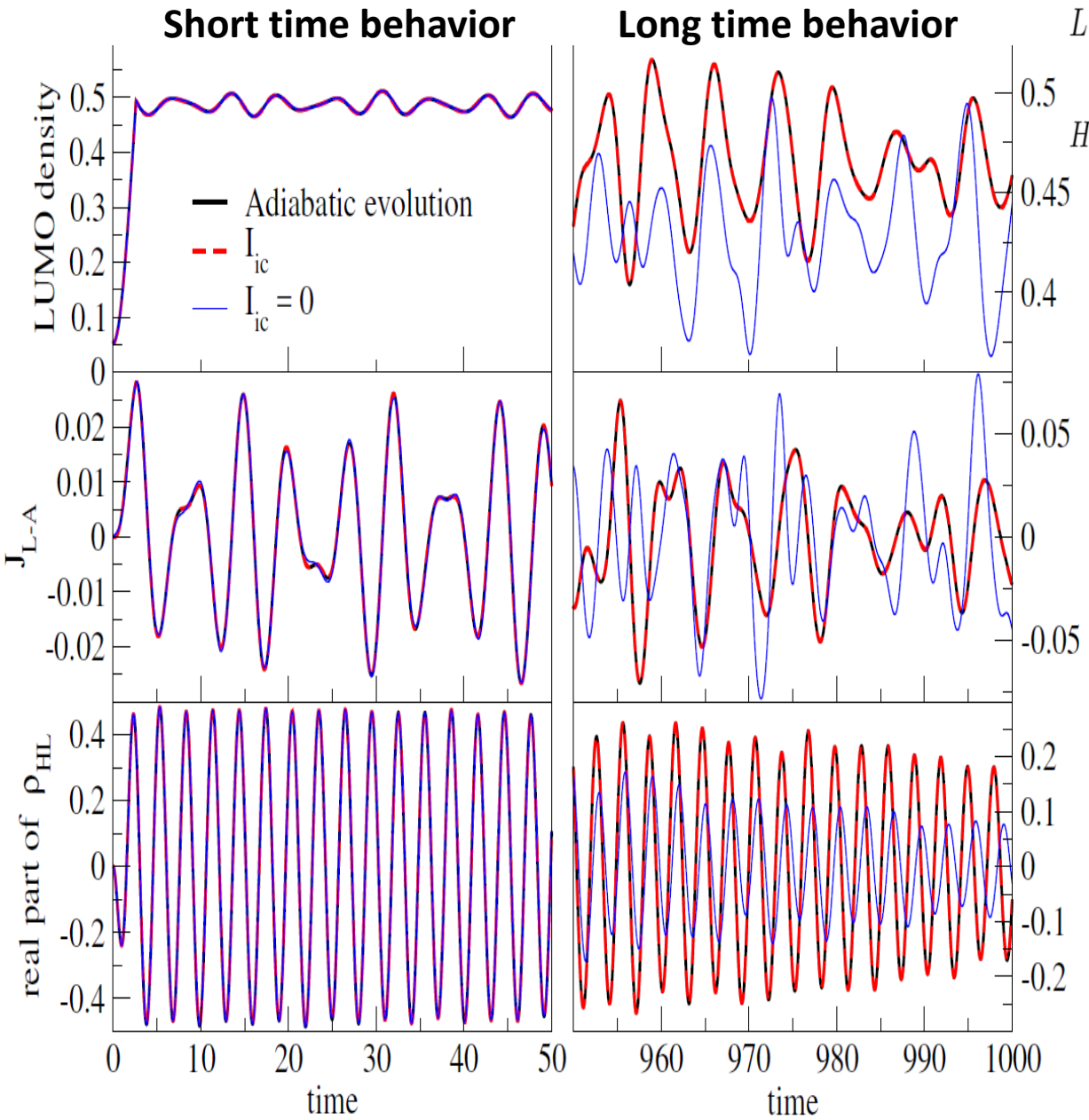
Density at LUMO without external fields

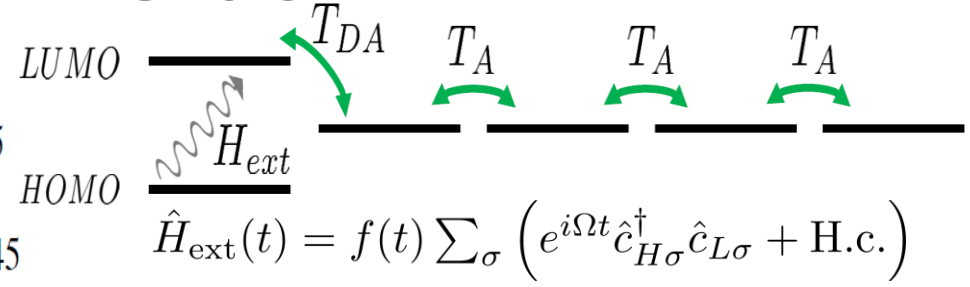
Initial correlations with external fields

Density at LUMO

current between LUMO-Acceptor

real part of density matrix between HOMO-LUMO





- Also off-diagonal parts of density matrix reproduced
- Initial correlations important at long times. Opposite behavior as found in homogeneous systems, where one can expect a decay in the initial correlations

  Semkat, D., Kremp, D., & Bonitz, M PRE **59**, 1557 (1999)

Conclusions

- ✓ Obtain expression for initial correlations independent on adiabatic switching
- ✓ Expressions valid without assuming losses, or homogenous systems
- ✓ Basis-independent expressions
- ✓ Separate the time propagation from the obtaining of the initial state
- ✓ Efficient implementation in existing GKBA codes

D. Karlsson, R. van Leeuwen, E. Perfetto, G. Stefanucci,
The generalized Kadanoff-Baym ansatz with initial correlations. PRB (2018)