# Time-dependent adiabatic *GW*
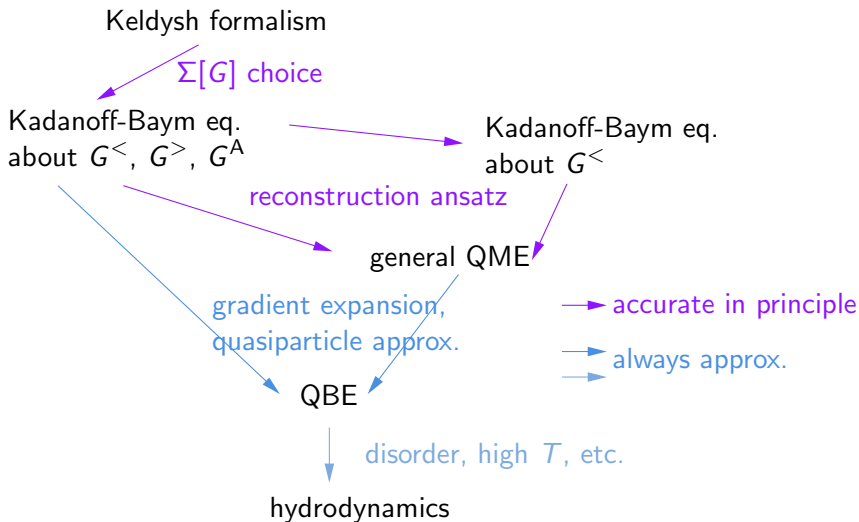
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# Relation between formalisms

**Motivation**

$$\langle A \rangle = \langle S^{-1} \mathcal{T}_t(S A_{\mathsf{I}}(t)) \rangle, \quad S = U(\infty, -\infty) \tag{1}$$

Non-equilibrium state: not pure; contains excited state components;
$|\Psi_n\rangle$ is excited state $\Rightarrow S|\Psi_n\rangle \neq e^{i\alpha}|\Psi_n\rangle \Rightarrow$ we can't peel the $S^{-1}$ off!!
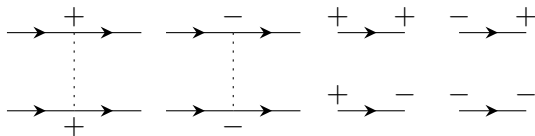
**Solution** Four (instead of one) types of propagators: (note $S^{-1}$ is *anti*-time ordered)
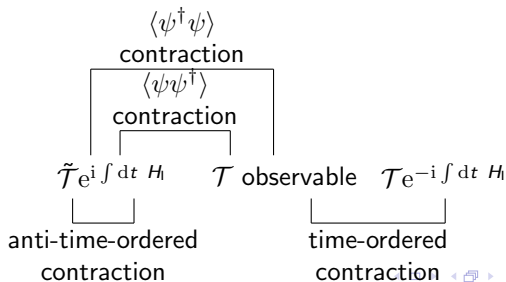
**Four types of (fermionic) propagators**

$$
\begin{aligned}
\mathrm{i}\, G^{--} &= \mathrm{i}\, G^{\mathsf{c}} = \langle \mathcal{T}\, \psi_1 \psi_2^{\dagger} \rangle, \quad \mathrm{i}\, G^{++} = \mathrm{i}\, G^{\mathsf{a}} = \langle \tilde{\mathcal{T}} \psi_1 \psi_2^{\dagger} \rangle, \\
\mathrm{i}\, G^{+-} &= \mathrm{i}\, G^{>} = \langle \psi_1 \psi_2^{\dagger} \rangle, \quad \mathrm{i}\, G^{-+} = \mathrm{i}\, G^{<} = -\langle \psi_2^{\dagger} \psi_1 \rangle.
\end{aligned}
\tag{2}
$$

**Diagrams**



**Self-energy**
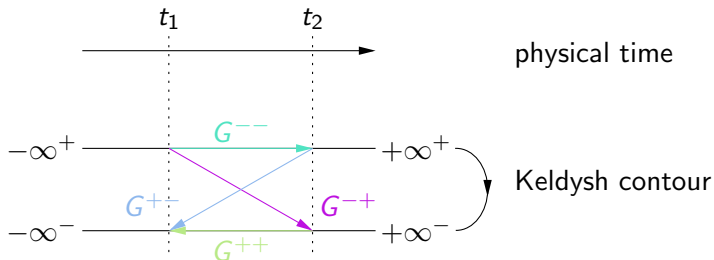
$$
G = \begin{pmatrix} G^{--} & G^{-+} \\ G^{+-} & G^{++} \end{pmatrix}, \quad \Sigma = \begin{pmatrix} \Sigma^{--} & \Sigma^{-+} \\ \Sigma^{+-} & \Sigma^{++} \end{pmatrix}, \quad G = G_0 + G_0 \Sigma G.
\tag{3}
$$

# Alternative formulation: Keldysh contour

**Keldysh contour** The information in the $G$ matrix can be alternatively stored in a time-ordered Green function on *Keldysh contour*

# Green function EOM

**From Keldysh contour to physical contour** Lengreth theorem:

$$(AB)^< = A^R B^< + A^< B^A, \quad (AB)^> = A^R B^> + A^> B^A,$$
$$(AB)^R = A^R B^R, \quad (AB)^A = A^A B^A, \tag{4}$$

where

$$A^>(t_1, t_2) = A(t_1^+, t_2^-), \quad A^<(t_1, t_2) = A(t_1^-, t_2^+),$$
$$A^R(t_1, t_2) = \theta(t_1 - t_2)(A^> - A^<), \tag{5}$$
$$A^A(t_1, t_2) = -\theta(t_1 - t_2)(A^> - A^<).$$

Mapping an equation on Keldysh contour to its counterpart on the physical time axis!

**Recommended references** The following series:

- Václav Špička, Bedřich Velický, and Anděla Kalvová. "Long and short time quantum dynamics: I. Between Green's functions and transport equations". In: *Physica E: Low-dimensional Systems and Nanostructures* 29.1-2 (2005), pp. 154–174

- Jørgen Rammer and H Smith. "Quantum field-theoretical methods in transport theory of metals". In: *Reviews of modern physics* 58.2 (1986), p. 323

**From self-energy correction to EOM** From Lengreth theorem:

$$G = G_0 + G_0 \Sigma G \Rightarrow G^< = G_0^< + G_0^< \Sigma^A G^A + G_0^R \Sigma^R G^< + G_0^R \Sigma^< G^A, \quad (6)$$

$$G = G_0 + G \Sigma G_0 \Rightarrow G^< = G_0^< + G_0^R \Sigma^R G_0^< + G^R \Sigma^< G_0^A + G^< \Sigma^A G^A, \quad (7)$$

$$G^A = G_0^A + G_0^A \Sigma^A G^A, \quad G^R = G_0^R + G_0^R \Sigma^R G^R. \quad (8)$$

**Getting rid of** $G_0$ We define

$$G_0^{-1} := i \partial_t - H_0, \quad (9)$$

and

$$G_0^{-1} G_0^{A,R} = I, \quad G_0^{-1} G_0^{<,>} = 0. \quad (10)$$

Taking complex conjugate of the def. of $G_0^{<,>}$ we find (left arrow = apply $\partial_t$ and $H_0$ to the second index of $G_0^{<,>}$)

$$G_0^{<,>} (-i \overleftarrow{\partial_{t_2}} - H_0) = 0. \quad (11)$$

**The Schrödinger-like Kadanoff-Baym eq.** Applying $G_0^{-1}$ to the left of (6) and to the right of (7):

$$(\mathrm{i}\,\partial_{t_1} - H_0)G^<(1,2) = \Sigma^R G^< + \Sigma^< G^A, \tag{12}$$

$$-\mathrm{i}\,\partial_{t_2} G^<(1,2) - G^< H_0 = G^R \Sigma^< + G^< \Sigma^A, \tag{13}$$

$$\Rightarrow \mathrm{i}(\partial_{t_1} + \partial_{t_2})G^< - [H_0, G^<] = \Sigma^R G^< + \Sigma^< G^A - G^R \Sigma^< - G^< \Sigma^A. \tag{14}$$

**Mixed coordinates** We define "average time" and "relative time":

$$T = \frac{t_1 + t_2}{2}, \quad t = t_1 - t_2, \tag{15}$$

$$\Rightarrow \frac{\partial}{\partial T} = \frac{\partial}{\partial t_1} + \frac{\partial}{\partial t_2}. \tag{16}$$

We then do Fourier transform over $t$: similar to the equilibrium case. ($T \simeq$ driving, $t \simeq$ internal time evolution)

# Towards a single-time formalism

**Summary up to now**

- *Accurate* EOMs about $G^{A,R}$, and EOM of $G^<$:

$$\mathrm{i}\,\partial_T G^< - [H_0, G^<] = \Sigma^R G^< + \Sigma^< G^A - G^R \Sigma^< - G^< \Sigma^A. \qquad (17)$$

  The RHS contains $t$ (or $\omega$) and $G^<$.

- Note: we can actually put the $t = 0$ part of $\Sigma$ into $H_0$! $\Rightarrow$ Example: COHSEX TD-aGW

**Goal** Obtaining quantum kinetics:

- Quantum master equation (QME), i.e. EOM of $\rho(\boldsymbol{r}_1, \boldsymbol{r}_2, t)$,
- and its downfolded version (in long wave length, well-defined quasiparticle limit), the quantum Boltzmann equation (QBE)

**Problem** Both LHS and RHS contain $\omega$: problem too large.
**What we want** Obtaining a close form EOM about $G^<(T, t = 0)$

# Quantum master equation

**Reduced density matrix** Single-electron density matrix:

$$i\,\rho(T) = G^<(T, t = 0) = \int \frac{d\omega}{2\pi} G^<(T, \omega) \tag{18}$$

**What we want** Two types of reduction:

- Reducing $\Sigma$ to an easy function of $G$, ideally $G^<$
- Reducing $G^<$ to $\rho(T)$

**Reducing $\Sigma$**

- Always possible: we can formally eliminate $\chi, \epsilon$, etc. from Hedin eq. and get a $\Sigma$ about $G$ i.e. about $G^<, G^{A,R}$
- But then $G^{A,B}$ can be eliminated with (8) as well
- In reality: a truncation is needed ...

# Reconstruction of $G^<$ from $\rho$

**Reconstruction theorem** From $\rho$, $G^{A,R}$ (which can be calculated using (8) from $\rho$), $G^<$ can be completely restored[1]

**Constructive proof** See (71) in the reference; note that

$$
\begin{aligned}
(G^R)^{-1}\theta(t_1 - t_2)G^< &= (\partial_{t_1} - H_0 - \Sigma^R)\theta(t_1 - t_2)G^< \\
&= \delta(t_1 - t_2)G^< + \theta(t_1 - t_2)(\partial_{t_1} - H_0 - \Sigma^R)G^< \\
&= \rho(t_1) + \cdots
\end{aligned}
\tag{19}
$$

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[1]Václav Špička, Bedřich Velický, and Anděla Kalvová. "Long and short time quantum dynamics: I. Between Green's functions and transport equations". In: *Physica E: Low-dimensional Systems and Nanostructures* 29.1-2 (2005), pp. 154–174

**Existence of accurate quantum master equation** In conclusion, in principle we can always write down something accurate like this:

$$\frac{\partial \rho}{\partial t} + i[H_0, \rho] = \int_{-\infty}^{t} F[\rho(t')] \, dt', \tag{20}$$

where $F$ is obtained from $\Sigma^R G^< + \Sigma^< G^A - G^R \Sigma^< - G^< \Sigma^A$, and $G^{R,A}$ is reconstructed from $\rho$ by doing a complete self-energy run, and $G^<$ is reconstructed from $G^A$ and $G^R$ and $\rho$.

**. . . but of course simplification is needed**

# Gradient expansion: first step from QME to QBE

**Mixed coordinates**

$$\tilde{\rho}(\boldsymbol{p}, \boldsymbol{X}, t) = \int \mathrm{d}x \, \mathrm{e}^{-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{x}} \, \rho\left(\boldsymbol{X} + \frac{\boldsymbol{x}}{2}, \boldsymbol{X} - \frac{\boldsymbol{x}}{2}, t\right), \qquad (21)$$

$$\frac{1}{\mathrm{i}}\widetilde{[H_0, \rho]} = \frac{\partial \epsilon}{\partial \boldsymbol{p}} \cdot \frac{\partial \tilde{\rho}}{\partial \boldsymbol{X}} - \frac{\partial \epsilon}{\partial \boldsymbol{X}} \cdot \frac{\partial \tilde{\rho}}{\partial \boldsymbol{p}} + \cdots \qquad (22)$$

**Gradient expansion** Only take the first two terms: assuming no higher dependence

# Issue: the definitions of $G_0$ and $\Sigma$

**Ambiguity in the meaning of $\Sigma$**

- In ordinary usage: $G_0$ directly from $H_0$
- But some prefer to move a part of $\Sigma$ that looks like "effective potential" into $H_0$ . . .
- $G_0$ contains "interactively corrected band structure"; $\Sigma$ contains "scattering" – what is the distinction?

**Comparison with similar issue in QBE**

- When impurities are rare: they appear in collision integral
- When impurities are abundant: they lead to an impurity band . . . and appear in the diffusion term?
- In QBE: it depends on the shape of the spectral function . . .

**Lacking proof of equivalence**

- Do different division of labor between $\Sigma$ and $G_0$ lead to consistent results?

**Approximations leading to QBE**

- Smooth $U_{\text{ext}} \Rightarrow$ Gradient expansion:

$$[H_0, \rho] \longrightarrow i\left(\frac{\partial \epsilon}{\partial \boldsymbol{p}} \cdot \frac{\partial \tilde{\rho}}{\partial \boldsymbol{X}} - \frac{\partial \epsilon}{\partial \boldsymbol{X}} \cdot \frac{\partial \tilde{\rho}}{\partial \boldsymbol{p}} + \cdots\right). \quad (23)$$

- Weakly correlated states $\Rightarrow$ Quasiparticle approx.:

$$G^<(\boldsymbol{X}, \boldsymbol{p}, T, \omega) = i\underbrace{2\pi\delta(\omega - \xi_{\boldsymbol{p}} + \mu - U(\boldsymbol{X}, T))}_{A(\boldsymbol{X}, \boldsymbol{p}, T, \omega)} f(\boldsymbol{p}, \boldsymbol{X}, T), \quad (24)$$

$$G^>(\boldsymbol{X}, \boldsymbol{p}, T, \omega) = -i\, A(\boldsymbol{X}, \boldsymbol{p}, T, \omega)(1 - f(\boldsymbol{p}, \boldsymbol{X}, T)). \quad (25)$$

This makes sense: we then have

$$A = i(G^R - G^A) = i(G^> - G^<). \quad (26)$$

- Gradient expansion in time domain $\Rightarrow$ Markovian collision integral

**Note**

- The conditions are sufficient, but not necessary: in the formalism above, field renormalization (as in electron-phonon interaction) is not included, but by correcting the collision term (essentially, a mild breakdown of Fermi golden rule), a Boltzmann equation can still be established (with necessary corrections).

- The first condition and the rest two conditions are orthogonal: the first condition can also be used in QME: it gives the diffusion part of QBE

- The second and third conditions are used to simply the interactive RHS into the collision integral

**Convolution in Green function EOM**

$$AB := \int \mathrm{d}2\, A(1,2)B(2,3). \tag{27}$$

**Gradient expansion, in $r$ and $t$** Taking Taylor expansion in $(\boldsymbol{r}, t)$

$$
\begin{aligned}
AB|_{\boldsymbol{X},\boldsymbol{p},T,\omega} = {} & A_{\boldsymbol{X},\boldsymbol{p},T,\omega} B_{\boldsymbol{X},\boldsymbol{p},T,\omega} \\
& + \frac{\mathrm{i}}{2}\left( \frac{\partial A}{\partial \boldsymbol{X}} \cdot \frac{\partial B}{\partial \boldsymbol{p}} - \frac{\partial A}{\partial \boldsymbol{p}} \cdot \frac{\partial B}{\partial \boldsymbol{X}} - \frac{\partial A}{\partial T}\frac{\partial B}{\partial \omega} + \frac{\partial A}{\partial \omega}\frac{\partial B}{\partial T} \right) + \cdots
\end{aligned} \tag{28}
$$

We only keep the terms shown above.
$\Rightarrow [G^<, H_0]$ reduces to the diffusion term seen in QBE

**Multi-band, spin index, etc.** When we have discrete labels in $A, B, \ldots$, quantities in (28) are matrices with these discrete indices

**Prototype of the collision integral** Keeping only the first term in gradient expansion (28):

$$
\begin{aligned}
&\Sigma^{R} G^{<} + \Sigma^{<} G^{A} - G^{R} \Sigma^{<} - G^{<} \Sigma^{A} \\
&\overset{\text{gradient exp.}}{\approx} G^{<}(\Sigma^{R} - \Sigma^{A}) - (G^{R} - G^{A})\Sigma^{<} \\
&= G^{<}\Sigma^{>} - G^{>}\Sigma^{<} \\
&\overset{\text{QP approx.}}{\approx} i\, A f(\boldsymbol{p}) \operatorname{Im} \Sigma - \text{incoming terms}
\end{aligned}
\tag{29}
$$

- Second line: the commutator terms assume the same form as that of (28), but are an order of magnitude smaller than it.
- Third line: from $A^{R} - A^{A} = A^{>} - A^{<}$
- The quasiparticle approximation (24) $\Rightarrow$ introduction of $f(\boldsymbol{p})$; the "out" part $\propto \operatorname{Im} \Sigma$ ($\operatorname{Im} \Sigma$ generally is even not well-defined)

**Obtaining the collision integral**

$$\text{LHS} = i\,\partial_T G^< - i\left(\frac{\partial H_0}{\partial \boldsymbol{X}} \cdot \frac{\partial G^<}{\partial \boldsymbol{p}} - \frac{\partial H_0}{\partial \boldsymbol{p}} \cdot \frac{\partial G^<}{\partial \boldsymbol{X}}\right), \tag{30}$$

where the $-\frac{\partial H_0}{\partial T}\frac{\partial G^<}{\partial \omega} + \frac{\partial H}{\partial \omega}\frac{\partial G^<}{\partial T}$ terms vanish when $H_0$ contains no strong time dependence.
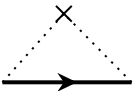
$$\text{RHS} = G^< \Sigma[i\,G^>] - G^> \Sigma[i\,G^<]. \tag{31}$$

$G^<$ appears in the LHS once, so there is a $2\pi\,i\,\delta(\omega - H)$ factor in the LHS; in the RHS suppose $\Sigma \sim G^n$, then the normalization factor coming with $G$ is $(2\pi\,i\,\delta(\omega - H))^{n+1}$; but since there are $n$ propagators in $\Sigma$, we have $n - 1$ frequency integrals connecting them to the "skeleton" of $\Sigma$, each with a factor of $1/2\pi$, so finally there is a $2\pi\delta(\omega - H)$ factor in the

RHS, which exactly is the factor seen in Fermi golden rule. The imaginary units always cancel each other finally.

**Example: disorder self-energy** The self-energy is

$$-i\,\Sigma(\boldsymbol{p},\omega) = \quad \underset{\longrightarrow}{\overset{\times}{\diagup\!\!\!\diagdown}} \quad = \sum_{\boldsymbol{q}} -\frac{1}{V}c|v(\boldsymbol{q}-\boldsymbol{p})|^2 \cdot i\,G(\boldsymbol{q},\omega), \quad (32)$$

where each dotted line corresponds to $-i\,gv(\boldsymbol{q}-\boldsymbol{p})/\sqrt{V}$, and $c$ comes from averaging over the distribution of disorders.

The equation becomes

$$
\begin{aligned}
&i\,\partial_T G^< - i\left(\frac{\partial H_0}{\partial \boldsymbol{X}} \cdot \frac{\partial G^<}{\partial \boldsymbol{p}} - \frac{\partial H_0}{\partial \boldsymbol{p}} \cdot \frac{\partial G^<}{\partial \boldsymbol{X}}\right) \\
&= \frac{1}{V}\sum_{\boldsymbol{q}} c|v(\boldsymbol{p}-\boldsymbol{q})|^2(i\,G^<(\boldsymbol{p}) \cdot G^>(\boldsymbol{q}) - i\,G^>(\boldsymbol{p}) \cdot G^<(\boldsymbol{q})),
\end{aligned}
\quad (33)
$$

and thus

# The coverage of quantum Boltzmann equation

- Exciton is multi-band phenomenon, but multi-band QBE can be established; see Lifshitz's Statistical Physics: Theory of the Condensed State, §5 for a magnetic field-induced exciton

- Plasmon comes from long-range divergence of Hartree term (in BerkeleyGW the $\boldsymbol{q} = 0$, $\boldsymbol{G} = 0$ part is omitted)

# A little beyond the quasiparticle approximation

When there *is* severe frequency dependence in $\Sigma$, but this has nothing to do with dissipation, we have QBE with field renormalization.[2]

**Example: electron-phonon interaction**  Renormalized QBE:

$$
\begin{aligned}
&(\partial_T + \boldsymbol{\nabla}_{\boldsymbol{p}} E_{\boldsymbol{p}} \cdot \boldsymbol{\nabla}_{\boldsymbol{R}} - \boldsymbol{\nabla}_{\boldsymbol{R}}(E_{\boldsymbol{p}} + e\varphi) \cdot \boldsymbol{\nabla}_{\boldsymbol{p}}) n_{\boldsymbol{p}} \\
&= -2\pi \int \frac{\mathrm{d}^3 \boldsymbol{p}'}{(2\pi)^3} Z_{\boldsymbol{p}} Z_{\boldsymbol{p}'} |g_{\boldsymbol{p}-\boldsymbol{p}'}|^2 \\
&\quad \times (n_{\boldsymbol{p}}(1 - n_{\boldsymbol{p}'})(1 + N(E_{\boldsymbol{p}} - E_{\boldsymbol{p}'})) - n_{\boldsymbol{p}'}(1 - n_{\boldsymbol{p}})N(E_{\boldsymbol{p}} - E_{\boldsymbol{p}'})) \\
&\quad \times (\delta(E_{\boldsymbol{p}} - E_{\boldsymbol{p}'} - \omega_{\boldsymbol{p}-\boldsymbol{p}'}) - \delta(E_{\boldsymbol{p}} - E_{\boldsymbol{p}'} + \omega_{\boldsymbol{p}-\boldsymbol{p}'}))
\end{aligned}
\tag{34}
$$

$\Rightarrow$ Fermi golden rule is not accurate when field renormalization is strong

**Another example: DMFT QBE**[3]

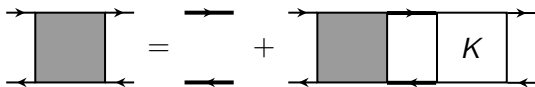[2] Jørgen Rammer and H Smith. "Quantum field-theoretical methods in transport theory of metals". In: *Reviews of modern physics* 58.2 (1986), p. 323.

[3] Michael Wais et al. "Quantum Boltzmann equation for strongly correlated systems: Comparison to dynamical mean field theory". In: *Physical Review B* 98.13 (2018), p. 134312.
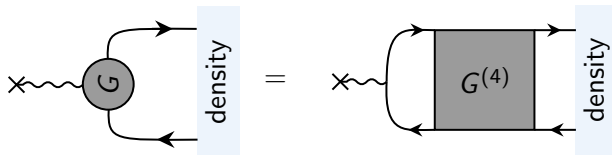
**BSE for second-order correlation**
Bethe–Salpeter equation (BSE)



$$\text{(35)}$$

**What we need** Linear response of single-electron under external field $=$ BSE (simplest single-electron theory: QBE)



$$\text{(36)}$$

**Next step: relation between $K$ and $\Sigma$**

**Linear response of a single self-energy diagram**



$$\Sigma = \quad \xrightarrow{\text{driving}} \quad \xrightarrow{\text{linear res.}} \qquad (37)$$

**Whole picture**



$$(38)$$
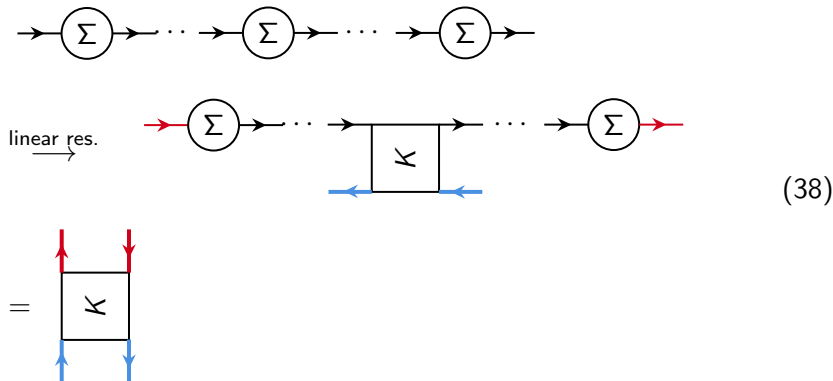
**Example: linear response from time-dependent $GW = $ BSE**



$$\Sigma = \quad\bigcirc\quad + \quad\overset{\frown}{\longrightarrow}\quad , \tag{39}$$

$$K = \quad \rangle\cdots\cdots\langle \quad + \quad \Vert$$

- First term = Electron Hartree term = Electron direct term = Exciton exchange term; $+1$ prefactor;
- Second term = Electron Fock term = Electron exchange term = Exciton direct term; $(-1)$ prefactor.

# Summary of formalisms

- Keldysh formalism (TODO: subtleties in initial correlation)
  - In principle we can get a closed equation system (with retardation) about $G$ and hence $G^<$;
  - in practice $\Sigma[G]$ has to be truncated; $n$ corrected propagator in $\Sigma = n$-order non-trivial correlation
- $\Rightarrow$ ... and hence a (highly complicated) accurate quantum master equation
  - $G$ needs to be reconstructed from $\rho$: reconstruction formalism
  - Issue: how to decide the division of labor between $H_0$ and $\Sigma$, when no physical pictures like "distinction between diffusion and collision" are available?
- Boltzmann formalism
  - Approximation 1: gradient expansion
  - Approximation 2: well-defined quasiparticle
  - Slight violation of approx. 2 (e.g. $Z$ renormalization factor): Fermi golden rule no longer 100 % correct
- QBE $\Rightarrow$ hydrodynamics with random fluctuation

# Time-dependent adiabatic *GW*

**Overview**

- ("Adiabatic") approximation for $\Sigma$: static limit of *GW* (i.e. $t = 0$) $=:$ static COHSEX
- In linear limit: $W$ doesn't change $\Rightarrow$ only high-order correlation taken into account is the ladder approximation using static screening $=$ static screening BSE
- $\Sigma$ has no $t \neq t'$ components $\Rightarrow$ $\Sigma$ can be placed into $H_0 \Rightarrow$ TD-aGW usually carried out in QME framework

# Introduction to COHSEX

# Example: