

Time-dependent adiabatic GW

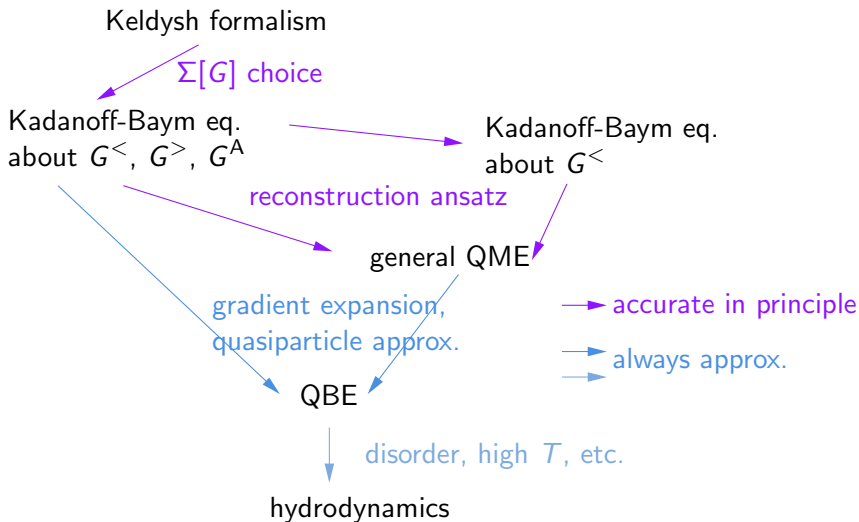
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Relation between formalisms



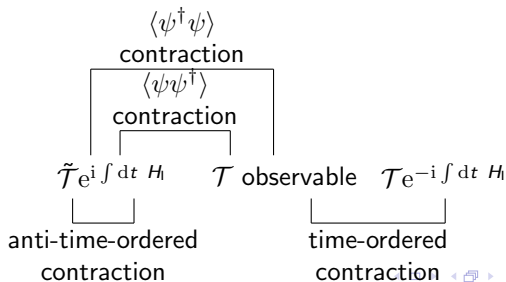
Non-equilibrium Green function

Motivation

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

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!!

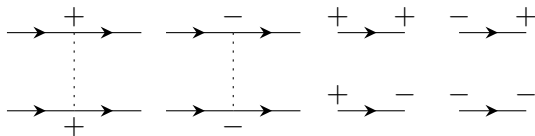
Solution Four (instead of one) types of propagators: (note S^{-1} is *anti*-time ordered)



Four types of (fermionic) propagators

$$\begin{aligned} iG^{--} &= iG^c = \langle \mathcal{T} \psi_1 \psi_2^\dagger \rangle, & iG^{++} &= iG^a = \langle \tilde{\mathcal{T}} \psi_1 \psi_2^\dagger \rangle, \\ iG^{+-} &= iG^> = \langle \psi_1 \psi_2^\dagger \rangle, & iG^{-+} &= iG^< = -\langle \psi_2^\dagger \psi_1 \rangle. \end{aligned} \quad (2)$$

Diagrams

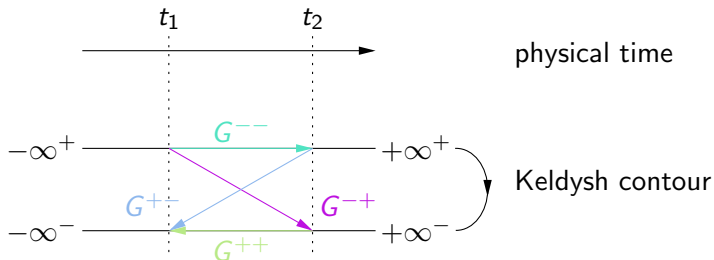


Self-energy

$$G = \begin{pmatrix} G^{--} & G^{-+} \\ G^{+-} & G^{++} \end{pmatrix}, \quad \Sigma = \begin{pmatrix} \Sigma^{--} & \Sigma^{-+} \\ \Sigma^{+-} & \Sigma^{++} \end{pmatrix}, \quad G = G_0 + G_0 \Sigma G. \quad (3)$$

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*



From Keldysh contour to physical contour Lengreth theorem:

$$\begin{aligned}(AB)^{<} &= A^R B^{<} + A^{<} B^A, & (AB)^{>} &= A^R B^{>} + A^{>} B^A, \\ (AB)^R &= A^R B^R, & (AB)^A &= A^A B^A,\end{aligned}\tag{4}$$

where

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

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

Derivation of EOM of $G^{<, >}$ and G^A I

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

Derivation of EOM of $G^{<,>}$ and G^A II

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 ∂_t and H_0 to the second index of $G_0^{<,>}$)

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

Derivation of EOM of $G^{<,>}$ and G^A III

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

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

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

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

Mixed coordinates We define “average time” and “relative time”:

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

$$\Rightarrow \frac{\partial}{\partial T} = \frac{\partial}{\partial t_1} + \frac{\partial}{\partial t_2}. \quad (16)$$

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 $G^<$:

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

The RHS contains t (or ω) and $G^<$.

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

Goal Obtaining quantum kinetics:

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

Problem Both LHS and RHS contain ω : 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:

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

What we want Two types of reduction:

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

Reducing Σ

- Always possible: we can formally eliminate χ, ϵ , etc. from Hedin eq. and get a Σ 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 ρ

Reconstruction theorem From ρ , $G^{A,R}$ (which can be calculated using (8) from ρ), $G^<$ can be completely restored¹

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

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

¹Václav 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. 

Quantum master equation as an accurate formalism

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

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

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

... but of course simplification is needed

Gradient expansion: first step from QME to QBE

Mixed coordinates

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

$$\frac{1}{i} \widetilde{[H_0, \rho]} = \frac{\partial \epsilon}{\partial \mathbf{p}} \cdot \frac{\partial \tilde{\rho}}{\partial \mathbf{X}} - \frac{\partial \epsilon}{\partial \mathbf{X}} \cdot \frac{\partial \tilde{\rho}}{\partial \mathbf{p}} + \dots \quad (22)$$

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

Issue: the definitions of G_0 and Σ

Ambiguity in the meaning of Σ

- In ordinary usage: G_0 directly from H_0
- But some prefer to move a part of Σ that looks like “effective potential” into H_0 ...
- G_0 contains “interactively corrected band structure”; Σ 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 Σ and G_0 lead to consistent results?

A radical move towards quantum Boltzmann equation I

Approximations leading to QBE

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

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

- Weakly correlated states \Rightarrow Quasiparticle approx.:

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

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

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

A radical move towards quantum Boltzmann equation II

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 simplify the interactive RHS ($G^>\Sigma^< - G^<\Sigma^>$) into the collision integral.

A radical move towards quantum Boltzmann equation III

- $\Sigma^{<,>}$ are intuitively related to scattering but scattering \neq non-pure state evolution (“incoherence”); Markovian also \neq dissipation (counterexample: quantum optics beam splitter, which has a scattering matrix and splits beam immediately – but is totally pure-state i.e. coherent). Incoherence only appears due to ignoring other degrees of freedom (and not using an input-output formalism), which comes from higher-order correlation being ignored here.

A radical move towards quantum Boltzmann equation IV

Convolution in Green function EOM

$$AB := \int d2 A(1, 2) B(2, 3). \quad (27)$$

Gradient expansion, in \mathbf{r} and t Taking Taylor expansion in (\mathbf{r}, t)

$$AB|_{\mathbf{X}, \mathbf{p}, T, \omega} = A_{\mathbf{X}, \mathbf{p}, T, \omega} B_{\mathbf{X}, \mathbf{p}, T, \omega} + \frac{i}{2} \left(\frac{\partial A}{\partial \mathbf{X}} \cdot \frac{\partial B}{\partial \mathbf{p}} - \frac{\partial A}{\partial \mathbf{p}} \cdot \frac{\partial B}{\partial \mathbf{X}} - \frac{\partial A}{\partial T} \frac{\partial B}{\partial \omega} + \frac{\partial A}{\partial \omega} \frac{\partial B}{\partial T} \right) + \dots \quad (28)$$

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, \dots , quantities in (28) are matrices with these discrete indices

A radical move towards quantum Boltzmann equation V

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

$$\begin{aligned}
 & \Sigma^R G^< + \Sigma^< G^A - G^R \Sigma^< - G^< \Sigma^A \\
 & \stackrel{\text{gradient exp.}}{\approx} G^< (\Sigma^R - \Sigma^A) - (G^R - G^A) \Sigma^< \\
 & = G^< \Sigma^> - G^> \Sigma^< \\
 & \underbrace{\stackrel{\text{QP approx.}}{\approx} \text{collision integral}}_{\text{molecular chaos hypothesis introduced}} \quad \stackrel{\text{relaxation time approx.}}{\approx} i A f(\mathbf{p}) \text{Im } \Sigma
 \end{aligned} \tag{29}$$

- 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 $f(\mathbf{p})$; the “out” part $\propto \text{Im } \Sigma$ ($\text{Im } \Sigma$ generally is even not well-defined)

A radical move towards quantum Boltzmann equation VI

Obtaining the collision integral

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

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.

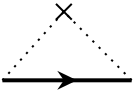
$$\text{RHS} = G^< \Sigma[i G^>] - G^> \Sigma[i G^<]. \quad (31)$$

$G^<$ 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 Σ , we have $n - 1$ frequency integrals connecting them to the “skeleton” of Σ , each with a factor of $1/2\pi$, so finally there is a $2\pi \delta(\omega - H)$ factor in the

A radical move towards quantum Boltzmann equation VII

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

$$-i\Sigma(\mathbf{p}, \omega) = \text{triangle diagram} = \sum_{\mathbf{q}} -\frac{1}{V} c |v(\mathbf{q} - \mathbf{p})|^2 \cdot iG(\mathbf{q}, \omega), \quad (32)$$


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

The equation becomes

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

A radical move towards quantum Boltzmann equation VIII

and thus from quasiparticle approximation (f and A also depend on \mathbf{X}, T)

$$\frac{d}{dT} A(\mathbf{p}) f(\mathbf{p}) = \int \frac{d^d \mathbf{q}}{(2\pi)^d} c |\mathbf{v}(\mathbf{p} - \mathbf{q})|^2 A(\mathbf{p}) f(\mathbf{p}) \cdot (-A(\mathbf{q})) (1 - f(\mathbf{q})) + \mathbf{p} \leftrightarrow \mathbf{q}, \quad (34)$$

and integrating over ω we get

$$\frac{d}{dT} f(\mathbf{p}) = \int \frac{d^d \mathbf{q}}{(2\pi)^d} c |\mathbf{v}(\mathbf{p} - \mathbf{q})|^2 (-2\pi) \delta(\epsilon_{\mathbf{p}} - \epsilon_{\mathbf{q}}) f(\mathbf{p}) (1 - f(\mathbf{q})) + \mathbf{p} \leftrightarrow \mathbf{q}, \quad (35)$$

which is exactly what is obtained by intuitively inserting Fermi golden rule to the RHS of QBE.

The role of the assumptions

- Gradient expansion does most of the heavy lifting job
- Quasiparticle approximation is used to write down an explicit collision integral
- The locality of the collision integral comes from gradient expansion on the temporal domain as well

Phenomenon covered

- 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 $\mathbf{q} = 0$, $\mathbf{G} = 0$ part is omitted)

A little beyond the quasiparticle approximation

ω dependence in Σ , no damping \Rightarrow QBE with field renormalization.²

Example: electron-phonon interaction Renormalized QBE:

$$\begin{aligned} & (\partial_T + \nabla_{\mathbf{p}} E_{\mathbf{p}} \cdot \nabla_{\mathbf{R}} - \nabla_{\mathbf{R}} (E_{\mathbf{p}} + e\varphi) \cdot \nabla_{\mathbf{p}}) n_{\mathbf{p}} \\ &= -2\pi \int \frac{d^3 \mathbf{p}'}{(2\pi)^3} Z_{\mathbf{p}} Z_{\mathbf{p}'} |g_{\mathbf{p}-\mathbf{p}'}|^2 \\ & \quad \times (n_{\mathbf{p}}(1 - n_{\mathbf{p}'})(1 + N(E_{\mathbf{p}} - E_{\mathbf{p}'})) - n_{\mathbf{p}'}(1 - n_{\mathbf{p}})N(E_{\mathbf{p}} - E_{\mathbf{p}'})) \\ & \quad \times (\delta(E_{\mathbf{p}} - E_{\mathbf{p}'} - \omega_{\mathbf{p}-\mathbf{p}'}) - \delta(E_{\mathbf{p}} - E_{\mathbf{p}'} + \omega_{\mathbf{p}-\mathbf{p}'})) \end{aligned} \quad (36)$$

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

Another example: DMFT QBE³

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

³Michael 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.

QBE in crystal

In crystal:

- If we work in \mathbf{r} representation, $u_{n\mathbf{k}}$ means the relation between \mathbf{r} and \mathbf{k} is not simply Fourier transform; we can't define $f(\mathbf{X}, \mathbf{p})$ directly from Wigner transform.
- If we work in \mathbf{k} representation, no well-defined \mathbf{r} is initially given.

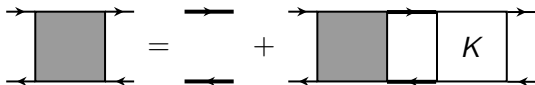
Solution

- Working in \mathbf{k} representation; expanding $\varepsilon_{n\mathbf{k}} + e\mathbf{r} \cdot \mathbf{E}$ in this basis;
- TODO: prove that $i\partial_{\mathbf{k}}$ is indeed the \mathbf{r} appearing in electric field; after Wigner transform in \mathbf{k} space, recover $\epsilon_{n\mathbf{k}} + e(\mathbf{r} + \mathbf{A}) \cdot \mathbf{E}$
- From semi-conservation law of \mathbf{k} we get localized collision integral in RHS; the matrix elements of the screened Coulomb interaction are evaluated in $\psi_{n\mathbf{k}}$ basis as well.

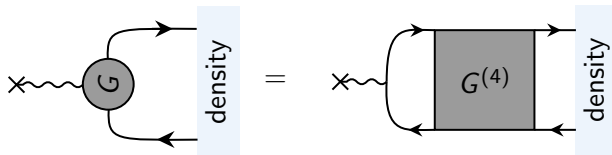
BSE and single-electron kinetic theory

BSE for second-order correlation

Bethe–Salpeter equation (BSE)


$$(37)$$

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


$$(38)$$

Next step: relation between K and Σ

Linear response of a single self-energy diagram

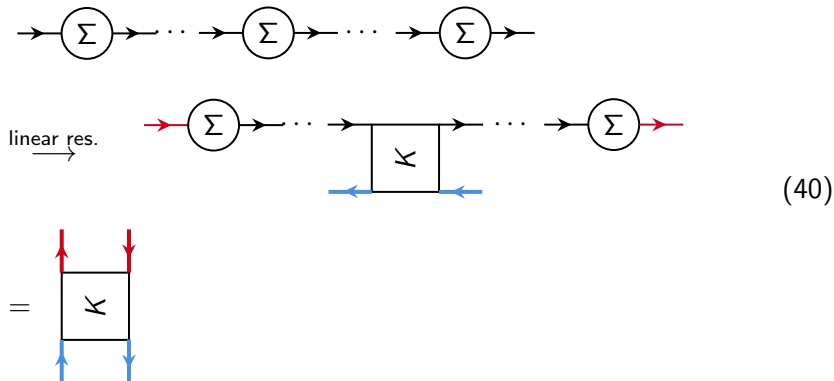
$$\Sigma = \text{[Diagram 1]} \xrightarrow{\text{driving}} \text{[Diagram 2]} \xrightarrow{\text{linear res.}} \text{[Diagram 3]} \quad (39)$$

The diagram illustrates the linear response of a single self-energy diagram Σ to an external driving force. It consists of three stages:



- Diagram 1:** A square box labeled K with two red arrows pointing right into and out of the top. A blue semi-circular arrow is attached to the bottom of the box, pointing clockwise.
- Diagram 2:** The same box K with red arrows on top. The blue semi-circular arrow is now connected to a wavy line with a cross at its end, representing an external driving force.
- Diagram 3:** The box K with two red arrows on top and two blue arrows on the bottom, all pointing right, representing a linear response.

Linking Σ with K


Whole picture



Example: linear response from time-dependent $GW = \text{BSE}$

$$\Sigma = \text{[Diagram 1]} + \text{[Diagram 2]}, \quad (41)$$



$$K = \text{[Diagram 3]} + \text{[Diagram 4]}$$



- 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
- $\Rightarrow \dots$ and hence a (highly complicated) accurate quantum master equation
 - G needs to be reconstructed from ρ : reconstruction formalism
 - Issue: how to decide the division of labor between H_0 and Σ , 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

Overview

- (“Adiabatic”) approximation for Σ : 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
- Σ 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: