

Time-dependent adiabatic GW

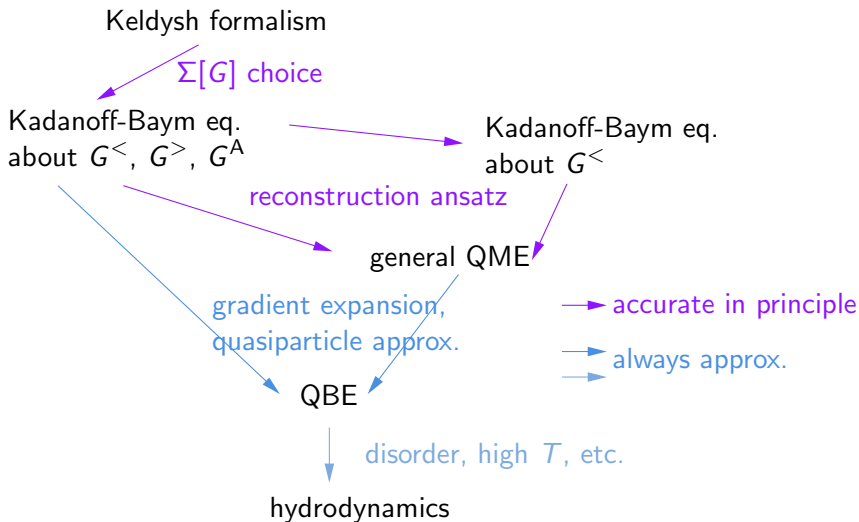
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Relation between formalisms



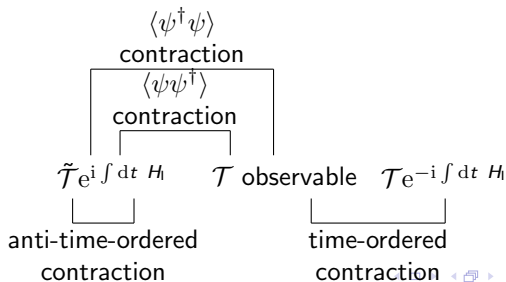
Non-equilibrium Green function

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!!

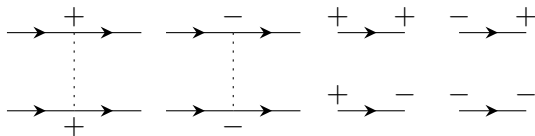
Solution Four (instead of one) types of propagators: (note S^{-1} is *anti*-time ordered)



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

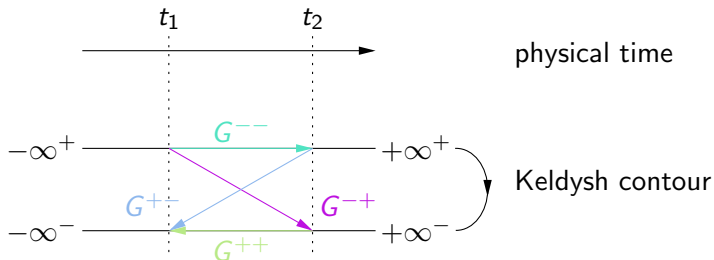


Self-energy

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

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

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

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

- Gradient expansion \Leftarrow smooth U_{ext} :

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

- Quasiparticle approx. \Leftarrow weak-correlated states:

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

- 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.
- 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 into the collision integral

A radical move towards quantum Boltzmann equation III

Convolution in Green function EOM

$$AB := \int d2 A(1, 2) B(2, 3). \quad (25)$$

Gradient expansion, in \mathbf{r} and t By def. and taking Taylor expansion in the (\mathbf{r}, t)

$$AB|_{\mathbf{x}, \mathbf{p}, T, \omega} = A_{\mathbf{x}, \mathbf{p}, T, \omega} B_{\mathbf{x}, \mathbf{p}, T, \omega} + \frac{i}{2} \left(\frac{\partial A}{\partial \mathbf{X}} \cdot \frac{\partial B}{\partial \mathbf{p}} - \frac{\partial A}{\partial \mathbf{p}} \cdot \frac{\partial B}{\partial \mathbf{X}} - \frac{\partial A}{\partial T} \frac{\partial B}{\partial \omega} + \frac{\partial A}{\partial \omega} \frac{\partial B}{\partial T} \right) + \dots \quad (26)$$

We only keep the terms shown above.

\Rightarrow hence the commutator $[G^<, H_0]$ can be reduced to the diffusion term seen in QBE

Multi-band, spin index, etc. When we have discrete labels in A, B, \dots , quantities in (26) are matrices with these discrete indices

Obtaining the collision integral

Keeping only the first term in gradient expansion (26):

$$\begin{aligned} & \Sigma^R G^< + \Sigma^< G^A - G^R \Sigma^< - G^< \Sigma^A \\ & \stackrel{\text{gradient exp.}}{\approx} G^< (\Sigma^R - \Sigma^A) - (G^R - G^A) \Sigma^< \\ & \stackrel{\text{QP approx.}}{\approx} i A f(\mathbf{p}) \text{Im } \Sigma - \text{incoming terms} \end{aligned} \quad (27)$$

Here in order to be consistent with the quasiparticle approximation of $G^<$, we also assume that $G^{A,R}$ assume the same forms as their equilibrium versions; thus the “out” part of the equation above $\propto \text{Im } \Sigma$ (in the most general non-equilibrium case $\text{Im } \Sigma$ is even not well-defined)

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

A little beyond the quasiparticle approximation

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

Example: electron-phonon interaction Renormalized QBE:

$$\begin{aligned} & (\partial_T + \nabla_{\mathbf{p}} E_{\mathbf{p}} \cdot \nabla_{\mathbf{R}} - \nabla_{\mathbf{R}} (E_{\mathbf{p}} + e\varphi) \cdot \nabla_{\mathbf{p}}) n_{\mathbf{p}} \\ &= -2\pi \int \frac{d^3 \mathbf{p}'}{(2\pi)^3} \mathbf{Z}_{\mathbf{p}} \mathbf{Z}_{\mathbf{p}'} |g_{\mathbf{p}-\mathbf{p}'}|^2 \\ & \quad \times (n_{\mathbf{p}}(1 - n_{\mathbf{p}'})(1 + N(E_{\mathbf{p}} - E_{\mathbf{p}'})) - n_{\mathbf{p}'}(1 - n_{\mathbf{p}})N(E_{\mathbf{p}} - E_{\mathbf{p}'})) \\ & \quad \times (\delta(E_{\mathbf{p}} - E_{\mathbf{p}'} - \omega_{\mathbf{p}-\mathbf{p}'}) - \delta(E_{\mathbf{p}} - E_{\mathbf{p}'} + \omega_{\mathbf{p}-\mathbf{p}'})) \end{aligned} \quad (28)$$

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

Another example: DMFT QBE³

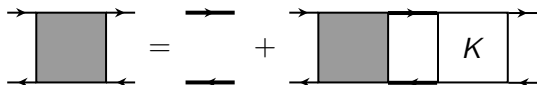
²Jørgen Rammer and H Smith. “Quantum field-theoretical methods in transport theory of metals”. In: *Reviews of modern physics* 58.2 (1986), p. 323.

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.

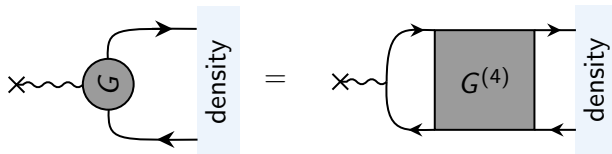
BSE and single-electron kinetic theory

BSE for second-order correlation

Bethe–Salpeter equation (BSE)


$$(29)$$

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


$$(30)$$

Next step: relation between K and Σ

Linear response of a single self-energy diagram

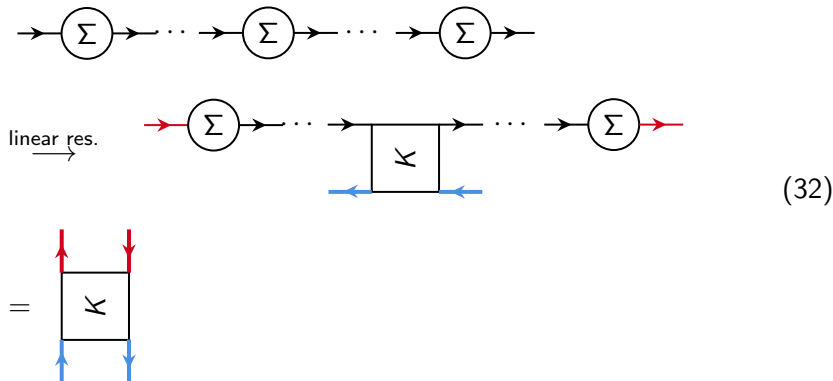
$$\Sigma = \text{[Diagram 1]} \xrightarrow{\text{driving}} \text{[Diagram 2]} \xrightarrow{\text{linear res.}} \text{[Diagram 3]} \quad (31)$$

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 $GW = \text{BSE}$

$$\Sigma = \text{[Diagram 1]} + \text{[Diagram 2]}, \quad (33)$$


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



- 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 ... 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: