Time-dependent adiabatic *GW*

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Non-equilibrium Green function

Motivation

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

Non-equilibrium state: not pure; contains excited state components; $|\Psi_n\rangle$ is excited state $\Rightarrow S\,|\Psi_n\rangle \neq \mathrm{e}^{\mathrm{i}\,\alpha}\,|\Psi_n\rangle \Rightarrow$ we can't peel the S^{-1} off!!

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

$\langle \psi^\dagger \psi \rangle$

contraction

$\langle \psi \psi^\dagger \rangle$

contraction

$\tilde{\mathcal{T}} \mathrm{e}^{\mathrm{i} \int \mathrm{d}t \; H_\mathrm{I}}$ $\quad \mathcal{T}$ observable $\quad \mathcal{T} \mathrm{e}^{-\mathrm{i} \int \mathrm{d}t \; H_\mathrm{I}}$

anti-time-ordered contraction

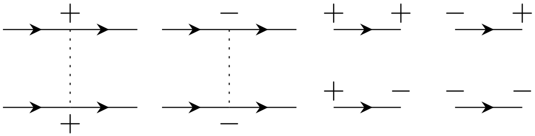
time-ordered contraction

Keldysh formalism

Four types of (fermionic) propagators

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

Diagrams

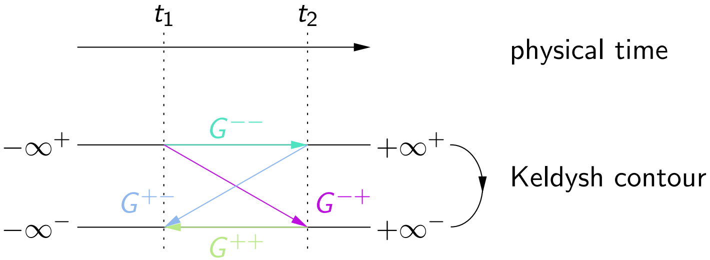


Self-energy

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

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:

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

where

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

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 Velickỳ, 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:

$$G = G_0 + G_0 \Sigma G \Rightarrow G^< = G_0^< + G_0^< \Sigma^{\mathrm{A}} G^{\mathrm{A}} + G_0^{\mathrm{R}} \Sigma^{\mathrm{R}} G^< + G_0^{\mathrm{R}} \Sigma^< G^{\mathrm{A}}, \tag{6}$$

$$G = G_0 + G \Sigma G_0 \Rightarrow G^< = G_0^< + G_0^{\mathrm{R}} \Sigma^{\mathrm{R}} G_0^< + G^{\mathrm{R}} \Sigma^< G_0^{\mathrm{A}} + G^< \Sigma^{\mathrm{A}} G^{\mathrm{A}}, \tag{7}$$

$$G^{\mathrm{A}} = G_0^{\mathrm{A}} + G_0^{\mathrm{A}} \Sigma^{\mathrm{A}} G^{\mathrm{A}}, \quad G^{\mathrm{R}} = G_0^{\mathrm{R}} + G_0^{\mathrm{R}} \Sigma^{\mathrm{R}} G^{\mathrm{R}}. \tag{8}$$

Getting rid of G_0 We define

$$G_0^{-1} := \mathrm{i}\,\partial_t - H_0, \tag{9}$$

and

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

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

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

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

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

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

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

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

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)

Summary up to now

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

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

  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 $\rho(\boldsymbol{r}_1, \boldsymbol{r}_2, t)$,
- and its long wave length 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:

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

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 $G^<, G^{\mathrm{A,R}}$
- But then $G^{\mathrm{A,B}}$ can be eliminated with (8) as well
- In reality: a truncation is needed ...

Reconstruction of $G^<$ from ρ

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

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

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

[1] Václav Špička, Bedřich Velickỳ, 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:

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

where F is obtained from $\Sigma^{\mathrm{R}} G^{<} + \Sigma^{<} G^{\mathrm{A}} - G^{\mathrm{R}} \Sigma^{<} - G^{<} \Sigma^{\mathrm{A}}$, and $G^{\mathrm{R,A}}$ 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

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

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

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 ...
- Thus: G_0 contains "interactively corrected band structure"; Σ contains "scattering"??

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?

Approximations leading to QBE

- Gradient expansion $\Leftarrow$ smooth U_{ext}:

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

- Quasiparticle approx. $\Leftarrow$ weak-correlated states:

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

- 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, mass 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 simply the interactive RHS into the collision integral

A radical move towards quantum Boltzmann equation III

Convolution in Green function EOM

$$AB := \int \mathrm{d}2\, A(1,2)B(2,3). \tag{25}$$

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

$$\begin{aligned} AB|_{\boldsymbol{X},\boldsymbol{p},T,\omega} &= A_{\boldsymbol{X},\boldsymbol{p},T,\omega} B_{\boldsymbol{X},\boldsymbol{p},T,\omega} \\ &+ \frac{\mathrm{i}}{2}\left(\frac{\partial A}{\partial \boldsymbol{X}} \cdot \frac{\partial B}{\partial \boldsymbol{p}} - \frac{\partial A}{\partial \boldsymbol{p}} \cdot \frac{\partial B}{\partial \boldsymbol{X}} - \frac{\partial A}{\partial T}\frac{\partial B}{\partial \omega} + \frac{\partial A}{\partial \omega}\frac{\partial B}{\partial T}\right) + \cdots \end{aligned} \tag{26}$$

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 $A, B, \ldots$, quantities in (26) are matrices with these discrete indices

Obtaining the collision integral

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

$$
\begin{aligned}
&\Sigma^{\mathrm{R}} G^{<} + \Sigma^{<} G^{\mathrm{A}} - G^{\mathrm{R}} \Sigma^{<} - G^{<} \Sigma^{\mathrm{A}} \\
\overset{\text{gradient exp.}}{\approx}\ & G^{<}(\Sigma^{\mathrm{R}} - \Sigma^{\mathrm{A}}) - (G^{\mathrm{R}} - G^{\mathrm{A}})\Sigma^{<} \\
\overset{\text{QP approx.}}{\approx}\ & \mathrm{i}\, A f(\boldsymbol{p}) \operatorname{Im}\Sigma -
\end{aligned}
\tag{27}
$$

Here from the quasiparticle approximation of $G^{<}$, we also assume that $G^{\mathrm{A,R}}$ assume the same forms as their equilibrium versions; thus the "out" part of the equation above $\propto \operatorname{Im}\Sigma$ (in the most general non-equilibrium case $\operatorname{Im}\Sigma$ 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 $\boldsymbol{q} = 0, \boldsymbol{G} = 0$ part is omitted)

Time-dependent adiabatic GW

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 $\Rightarrow$ Σ can be placed into H_0 $\Rightarrow$ TD-aGW usually carried out in QME framework

Introduction to COHSEX

Example: