Nonequilibrium Green functions simulations of femtosecond dynamics of quantum materials

Michael Bonitz, Jan-Philip Joost, Christopher Makait, Erik Schroedter, Hannes Ohldag, and Tim Kalsberger

Institute for Theoretical Physics and Astrophysics, Kiel University

in collaboration with Karsten Balzer (Kiel) and Iva Brezinova (TU Wien)

StatPhys, Lviv, August 2025, dedicated to the 100th birthday of Ihor Yukhnovskii

pdf of talk: https://www.itap.uni-kiel.de//theo-physik/bonitz/talks.html

Recent review: Bonitz et al., phys. stat. sol. (b) 2024, DOI: 10.1002/pssb.202300578

Recent review article. Credit

phys. stat. sol. (b) 261, 2300578 (2024)
"Accelerating Nonequilibrium Green Functions Simulations: The G1-G2 Scheme and Beyond"

Michael Bonitz, Jan-Philip Joost, Christopher Makait, Erik Schroedter, Tim Kalsberger, and Karsten Balzer

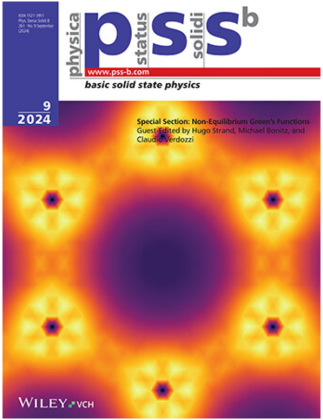


Fig.: G1-G2-simulation of laser excited graphene monolayer (Tim Kalsberger)

Jan-Philip Joost:
PhD 2023, NEGF simulations for graphene nanostructures, development of G1-G2 scheme

Motivation: Finite correlated quantum systems

Fermionic atoms in optical lattices
tunable lattice depth and interaction

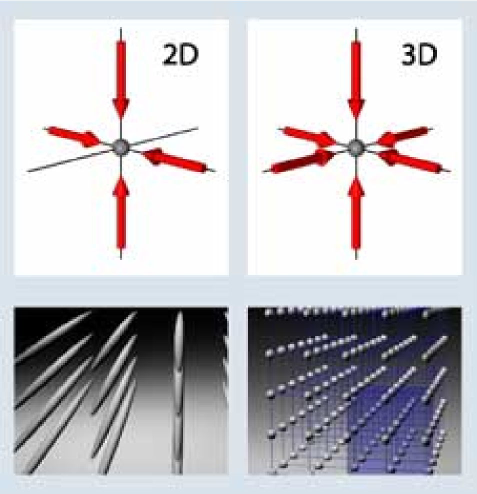
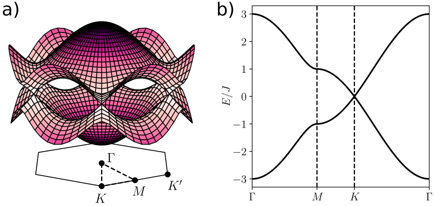
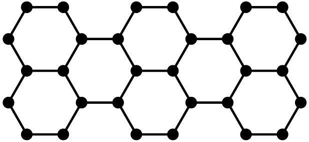


Fig.: M. Greiner (Harvard)

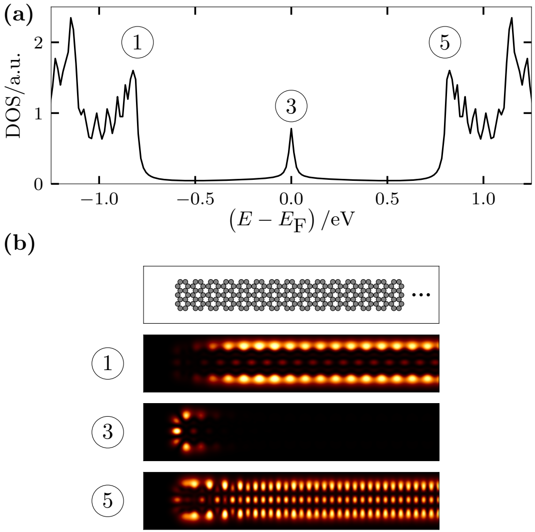
Graphene: high mobility, no bandgap



Alternatives : 1. TMDCs:
2. Graphene nanoribbons (GNR):
finite tunable bandgap, topological states



GNR: spatially localized spectral contributions[1]



- top: total density of states (DOS)

- DOS size and shape dependent

- many degrees of freedom: combination of two GNRs: heterostructures, combination of materials (TMDCs), multiple layers, twist angle
- bottom-up synthesis with atomic precision

- importance of e-e interactions

- what will happen in nonequilibrium, upon external excitation (e.g. by lasers or by ion impact)?

[1] 7 armchair GNR of 504 atoms, GW-NEGF ground state simulation,

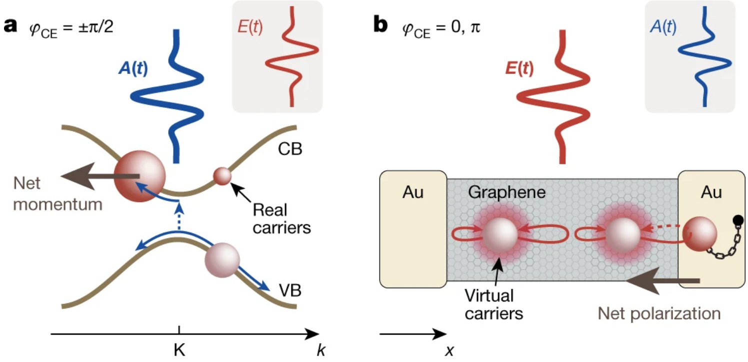
J.-P. Joost, A.-P. Jauho, and M. Bonitz, Nano Letters **19**, 9045 (2019)

Potential Applications of Graphene: towards petahertz electronics[2]

Experiments by P. Hommelhoff *et al.*: logic gate for lightwave electronics, variation of carrier envelope phase ϕ_{CE} of few cycle fs-laser pulse

a: momentum asymmetry $(A(t))$ creates $f_c(-k) \neq f_c(k)$ and net current

b: real space asymmetry $(E(t))$ of density creates net polarization



[2] Boolakee et al., Nature **605**, 251 (2022)

Time-dependent Schrödinger equation. Scaling bottleneck

- time-dependent many-electron Hamiltonian

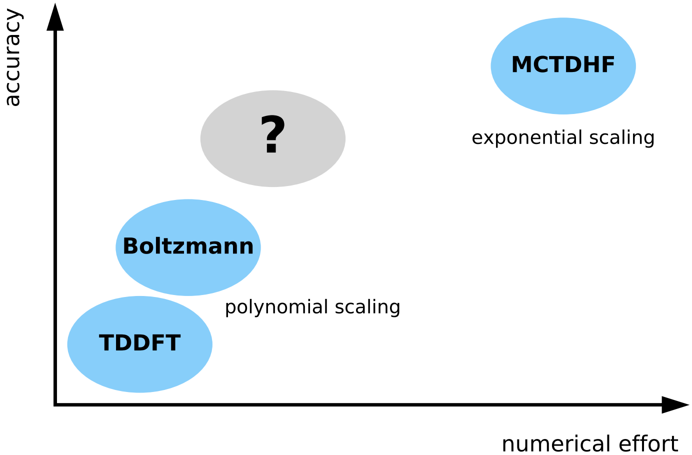
$$H(t) = \underbrace{\sum_{i=1}^{N} h(\boldsymbol{r}_i, t)}_{\text{one-body operators}} + \underbrace{\frac{1}{2}\sum_{i\neq j}^{N} W(\boldsymbol{r}_i, \boldsymbol{r}_j)}_{\textbf{pair-wise}\text{ interactions}}$$

- time-dependent Schrödinger equation (TDSE)

$$i\partial_t \Psi(\boldsymbol{r}_i, \ldots, \boldsymbol{r}_N; t) = H(t)\Psi(\boldsymbol{r}_i, \ldots, \boldsymbol{r}_N; t) \quad \cancel{\xrightarrow[\text{exponential scaling of numerical effort}]{\text{direct solution}}} \quad \Psi(\boldsymbol{r}_i, \ldots, \boldsymbol{r}_N; t)$$

- **solutions to overcome exponential scaling:**
  1. approximations to TDSE: TD-RASCI, TD-CASSCF, truncated CC, TD-R-matrix etc.
     D. Hochstuhl and M. Bonitz, PRA (2012) and EJP-ST (2014)
  2. propagation of simpler observables: density (TDDFT), distribution function (Kinetic theory), correlation functions etc.

Scaling of quantum many-body methods



*MCTDHF and other wavefunction-based methods (credit: I. Brezinova)

? Can one achieve sufficient accuracy (including e-e correlations) at non-exponential cost?

2nd quantization

- Fock space $\mathcal{F} \ni |n_1, n_2 \ldots\rangle$, $\mathcal{F} = \bigoplus_{N_0 \in \mathbb{N}} \mathcal{F}^{N_0}$, $\mathcal{F}^{N_0} \subset \mathcal{H}^{N_0}$
- $\hat{c}_i$,$\hat{c}_i^\dagger$ creates/annihilates a particle in single-particle orbital ϕ_i
- spin accounted for by canonical (anti-)commutator relations
  $\left[\hat{c}_i^{(\dagger)}, \hat{c}_j^{(\dagger)}\right]_\mp = 0, \quad \left[\hat{c}_i, \hat{c}_j^\dagger\right]_\mp = \delta_{i,j}$
- Hamiltonian: $\hat{H}(t) = \underbrace{\sum_{k,m} h^0_{km} \hat{c}_k^\dagger \hat{c}_m}_{\hat{H}_0} + \underbrace{\frac{1}{2} \sum_{k,l,m,n} w_{klmn} \hat{c}_k^\dagger \hat{c}_l^\dagger \hat{c}_n \hat{c}_m}_{\hat{W}} + \hat{F}(t)$

Particle interaction w_{klmn}

- Coulomb interaction
- electronic correlations

Time-dependent excitation $\hat{F}(t)$

- single-particle type
- em field, quench, particles

Nonequilibrium Green Functions (NEGF)

two times $z, z' \in \mathcal{C}$ ("Keldysh contour"), arbitrary one-particle basis $|\phi_i\rangle$

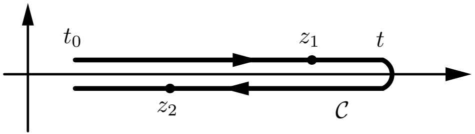
$$\boxed{G_{ij}(z,z') = \frac{\mathrm{i}}{\hbar}\left\langle \hat{T}_{\mathcal{C}} \hat{c}_i(z)\hat{c}_j^\dagger(z')\right\rangle}$$

average with $\hat{\rho}_N$

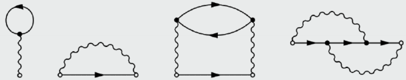
pure or mixed state

Keldysh–Kadanoff–Baym equations (KBE) on $\mathcal{C}$ (2×2 matrix)[3]:

$$\sum_k \left\{ \mathrm{i}\hbar \frac{\partial}{\partial z}\delta_{ik} - h_{ik}(z)\right\} G_{kj}(z,z') = \delta_{\mathcal{C}}(z,z')\delta_{ij} - \mathrm{i}\hbar \sum_{klm}\int_{\mathcal{C}} \mathrm{d}\bar{z}\, w_{iklm}(z^+,\bar{z}) G^{(2)}_{lmjk}(z,\bar{z};z',\bar{z}^+)$$



- $\int_{\mathcal{C}} wG^{(2)} \to \int_{\mathcal{C}} \Sigma G$, Selfenergy
- Nonequilibrium Diagram technique
  Example: Hartree–Fock + Second Born selfenergy



KBE: first equation of Martin–Schwinger hierarchy for $G, G^{(2)} \dots G^{(n)}$

[3] L.V. Keldysh ZhETF (1964), obituary: Bonitz, Jauho, Sadovskii, and Tikhodeev, phys. stat. sol. (b)(2019)

Real-Time Keldysh–Kadanoff–Baym Equations (KBE)

- Correlation functions $G^{\gtrless}$ obey real-time KBE

$$\sum_l \left[\mathrm{i}\hbar \frac{\mathrm{d}}{\mathrm{d}t} \delta_{i,l} - h^{\text{eff}}_{il}(t) \right] G^{>}_{lj}(t,t') = I^{(1),>}_{ij}(t,t') \,,$$

$$\sum_l G^{<}_{il}(t,t') \left[-\mathrm{i}\hbar \frac{\overleftarrow{\mathrm{d}}}{\mathrm{d}t'} \delta_{l,j} - h^{\text{eff}}_{lj}(t') \right] = I^{(2),<}_{ij}(t,t') \,,$$

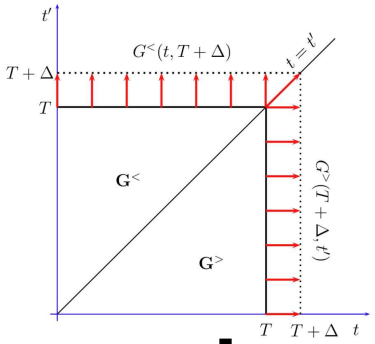
with the effective single-particle **Hartree–Fock Hamiltonian**

$$h^{\text{eff}}_{ij}(t) = h^0_{ij} \pm \mathrm{i}\hbar \sum_{kl} w^{\pm}_{ikjl} G^{<}_{lk}(t)$$

and the collision integrals

$$I^{(1),>}_{ij}(t,t') := \sum_l \int_{t_\mathrm{s}}^{\infty} \mathrm{d}\bar{t} \left\{ \Sigma^{\mathrm{R}}_{il}(t,\bar{t}) G^{>}_{lj}(\bar{t},t') + \Sigma^{>}_{il}(t,\bar{t}) G^{\mathrm{A}}_{lj}(\bar{t},t') \right\} ,$$

$$I^{(2),<}_{ij}(t,t') := \sum_l \int_{t_\mathrm{s}}^{\infty} \mathrm{d}\bar{t} \left\{ G^{\mathrm{R}}_{il}(t,\bar{t}) \Sigma^{<}_{lj}(\bar{t},t') + G^{<}_{il}(t,\bar{t}) \Sigma^{\mathrm{A}}_{lj}(\bar{t},t') \right\} .$$



$\longrightarrow \mathcal{O}(N_\mathrm{t}^3)$

- two-time structure contains spectral information
- numerically demanding due to cubic scaling with number of time steps N_t

Selfenergy Approximations[4]

Choice depends on coupling strength, density (filling)

Hartree–Fock (HF, mean field): $\sim w^1$

Second Born (2B): $\sim w^2$

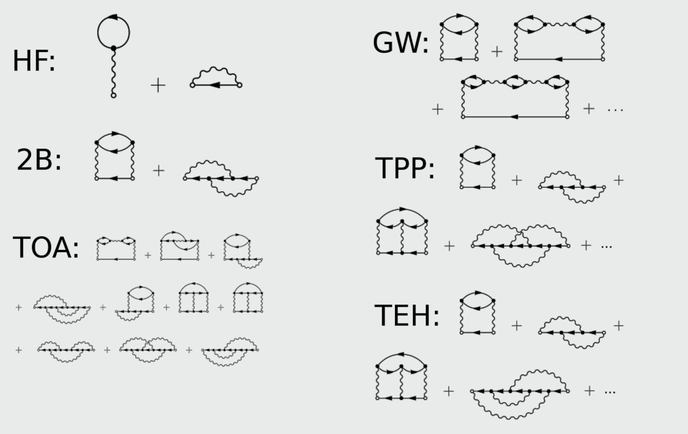
GW: ∞ bubble summation,
dynamical screening effects

particle-particle T-matrix (TPP):
∞ ladder sum in pp channel

particle-hole T-matrix (TPH/TEH):
∞ ladder sum in ph channel

3rd order approx. (TOA): $\sim w^3$

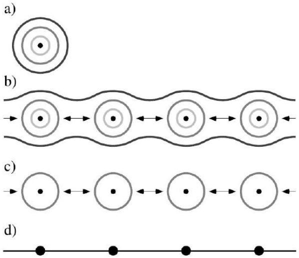
dynamically screened ladder (DSL)*:
$\sim \mathrm{2B} + GW + \mathrm{TPP} + \mathrm{TPH}$



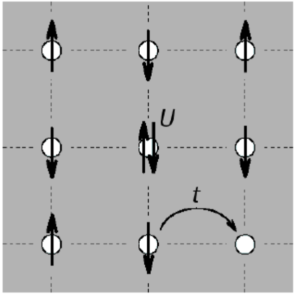
[4]Conserving approximations, nonequilibrium $\Sigma(t, t')$, applies for ultra-short to long times
Review: Schlünzen *et al.*, J. Phys. Cond. Matt. **32**, 103001 (2020); *Joost *et al.*, PRB **105** (2022)

The Hubbard model: accurate treatment of electronic correlations

- Simple, but versatile model for strongly correlated solid state systems, 2D quantum materials
- Suitable for single band, small bandwidth; atoms in optical lattices



$\Rightarrow$



$$\boxed{\hat{H}(t) = J\sum_{ij,\,\alpha} h_{ij}\,\hat{c}^\dagger_{i\alpha}\hat{c}_{j\alpha} + U\sum_i \hat{c}^\dagger_{i\uparrow}\hat{c}_{i\uparrow}\hat{c}^\dagger_{i\downarrow}\hat{c}_{i\downarrow} + \sum_{ij,\alpha\beta} f_{ij,\alpha\beta}(t)\,\hat{c}^\dagger_{i\alpha}\hat{c}_{j\beta}}$$

$h_{ij} = -\delta_{\langle i,\,j\rangle}$ and $\delta_{\langle i,\,j\rangle} = 1$, if (i,j) is nearest neighbor, $\delta_{\langle i,\,j\rangle} = 0$ otherwise

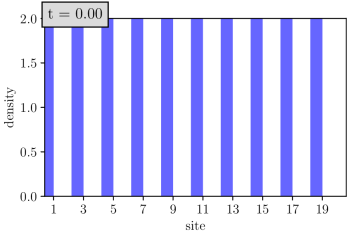
use $J = 1$, on-site repulsion $(U > 0)$ or attraction $(U < 0)$, tunable interaction strength

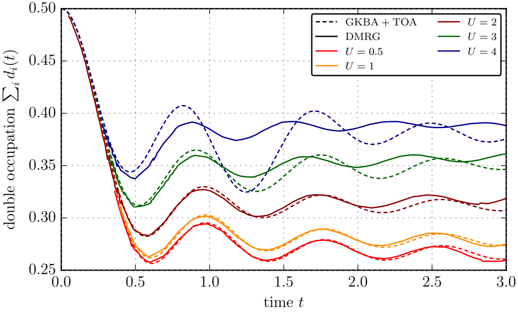
- parameters from electronic structure calculations or experiment
- systematic improvements: extended Hubbard and PPP model [Joost *et al.*, phys. stat. sol. (b) 2019]

Benchmarks of NEGF against DMRG (1D)[6]

Initial state:

charge density wave



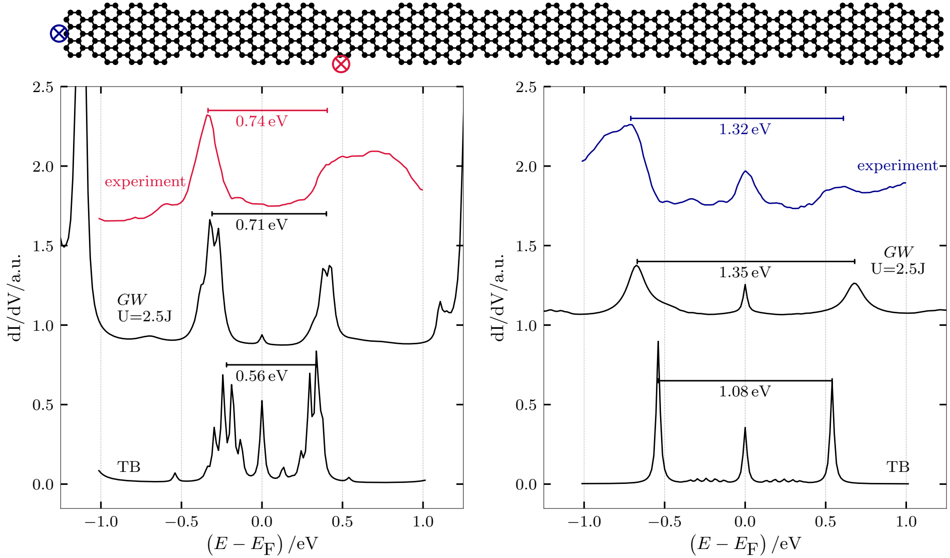


- sensitive observable: total double occupation
- good quality transients NEGF up to $U \simeq$ bandwidth
- accurate long-time behavior of GKBA+T-matrix (not shown)
- performance of different selfenergies vs. coupling and filling[5]

[5] N. Schlünzen, S. Hermanns, M. Scharnke, and M. Bonitz, J. Phys.: Cond. Matt. 32 (10), 103001 (2020)

[6] N. Schlünzen, J.-P. Joost, F. Heidrich-Meisner, and M. Bonitz, Phys. Rev. B **95**, 165139 (2017)

Local density of states of graphene nanoribbons: Bulk vs. End



Failure of tight binding and Hartree-Fock results. Electronic correlations crucial

Experiments: Rizzo et al. Nature, **560**, 204 (2018), NEGF simulations: Joost et al. Nano Lett. **19**, 9045 (2019)

Acceleration: Generalized Kadanoff–Baym Ansatz (GKBA)[7]

C | A | U
Christian-Albrechts-Universität zu Kiel
Mathematisch-Naturwissenschaftliche Fakultät

- full propagation on the time diagonal ($I := I^{(1),<}$):

$$i\hbar \frac{\mathrm{d}}{\mathrm{d}t} G^<_{ij}(t) = \left[h^{\mathrm{HF}}, G^<\right]_{ij}(t) + \left[I + I^\dagger\right]_{ij}(t)$$

- reconstruct off-diagonal NEGF from time diagonal:

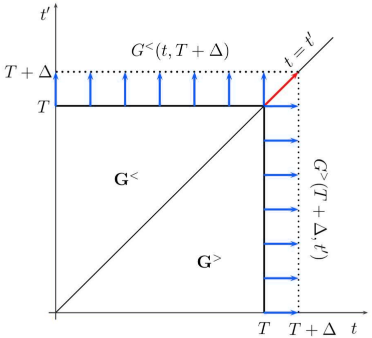
$$G^{\gtrless}_{ij}(t,t') = \pm\left[G^{\mathrm{R}}_{ik}(t,t')\rho^{\gtrless}_{kj}(t') - \rho^{\gtrless}_{ik}(t)G^{\mathrm{A}}_{kj}(t,t')\right]$$

with $\quad \rho^{\gtrless}_{ij}(t) = \pm i\hbar G^{\gtrless}_{ij}(t,t)$

- HF-GKBA: use Hartree–Fock propagators for $G^{\mathrm{R/A}}_{ij}$

$$G^{\mathrm{R/A}}_{ij}(t,t') = \mp i\Theta_{\mathcal{C}}\left(\pm[t-t']\right) \exp\left(-\frac{i}{\hbar}\int_{t'}^{t} \mathrm{d}\bar{t}\, h_{\mathrm{HF}}(\bar{t})\right)\Bigg|_{ij}$$

- conserves total energy
- Large number of applications to atoms, molecules, condensed matter systems, plasmas



$$\mathcal{O}(N_{\mathrm{t}}^2)$$

[6] P. Lipavský, V. Špička, and B. Velický, Phys. Rev. B **34**, 6933 (1986);
K. Balzer and M. Bonitz, Lecture Notes in Physics **867** (2013)

Reformulating the GKBA

CAU
Christian-Albrechts-Universität zu Kiel
Mathematisch-Naturwissenschaftliche Fakultät

- quadratic/cubic scaling is caused by the structure of the collision integral

$$I_{ij}(t) = \sum_k \underbrace{\boxed{\int_{t_0}^{t} \mathrm{d}\bar{t}}}_{\text{time integral}} \left[\Sigma^{>}_{ik}\boxed{(t,\bar{t})}G^{<}_{kj}\boxed{(\bar{t},t)} - \Sigma^{<}_{ik}\boxed{(t,\bar{t})}G^{>}_{kj}\boxed{(\bar{t},t)}\right]$$

off-diagonal functions

Reformulating the GKBA

- quadratic/cubic scaling is caused by the structure of the collision integral

$$I_{ij}(t) = \sum_k \underbrace{\boxed{\int_{t_0}^{t} \mathrm{d}\bar{t}}}_{\text{time integral}} \left[\Sigma^{>}_{ik}\boxed{(t,\bar{t})}G^{<}_{kj}\boxed{(\bar{t},t)} - \Sigma^{<}_{ik}\boxed{(t,\bar{t})}G^{>}_{kj}\boxed{(\bar{t},t)}\right] =: \pm\mathrm{i}\hbar \sum_{klp} w_{iklp}(t)\boxed{\mathcal{G}_{lpjk}(t)}$$

off-diagonal functions

Idea: solve differential equation for $\mathcal{G}$ instead of time integral for I

Reformulating the GKBA

- quadratic/cubic scaling is caused by the structure of the collision integral

$$I_{ij}(t) = \sum_k \underbrace{\boxed{\int_{t_0}^{t}} }_{\text{time integral}} \mathrm{d}\bar{t} \left[\Sigma^{>}_{ik}\boxed{(t,\bar{t})}G^{<}_{kj}\boxed{(\bar{t},t)} - \Sigma^{<}_{ik}\boxed{(t,\bar{t})}G^{>}_{kj}\boxed{(\bar{t},t)}\right] =: \pm \mathrm{i}\hbar \sum_{klp} w_{iklp}(t)\boxed{\mathcal{G}_{lpjk}(t)}$$

off-diagonal functions

Idea: solve differential equation for $\mathcal{G}$ instead of time integral for I

- example for 2B selfenergy[8]

$$\Sigma^{\gtrless}_{ij}\left(t,t'\right) = \pm\left(\mathrm{i}\hbar\right)^2 \sum_{klpqrs} w_{iklp}\left(t\right) w^{\pm}_{qrjs}\left(t'\right) G^{\gtrless}_{lq}\left(t,t'\right) G^{\gtrless}_{pr}\left(t,t'\right) G^{\lessgtr}_{sk}\left(t',t\right)$$

- respective $\mathcal{G}$ can be identified as

$$\mathcal{G}_{ijkl}(t) = \mathrm{i}\hbar \sum_{pqrs} \int_{t_0}^{t} \mathrm{d}\bar{t}\, w^{\pm}_{pqrs}\left(\bar{t}\right) \left[\mathcal{G}^{\mathrm{H},>}_{ijpq}(t,\bar{t})\mathcal{G}^{\mathrm{H},<}_{rskl}(\bar{t},t) - \mathcal{G}^{\mathrm{H},<}_{ijpq}(t,\bar{t})\mathcal{G}^{\mathrm{H},>}_{rskl}(\bar{t},t)\right]$$

with the two-particle Hartree Green function

$$\mathcal{G}^{\mathrm{H},\gtrless}_{ijkl}(t,t') := G^{\gtrless}_{ik}(t,t')G^{\gtrless}_{jl}(t,t')$$

[8] N. Schlünzen, J.-P. Joost and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020)

- quadratic/cubic scaling is caused by the structure of the collision integral

$$I_{ij}(t) = \sum_k \underbrace{\boxed{\int_{t_0}^{t} \mathrm{d}\bar{t}}}_{\text{time integral}} \left[\Sigma^{>}_{ik}\boxed{(t,\bar{t})}G^{<}_{kj}\boxed{(\bar{t},t)} - \Sigma^{<}_{ik}\boxed{(t,\bar{t})}G^{>}_{kj}\boxed{(\bar{t},t)}\right] =: \pm \mathrm{i}\hbar \sum_{klp} w_{iklp}(t)\boxed{\mathcal{G}_{lpjk}(t)}$$

off-diagonal functions

Idea: solve differential equation for $\mathcal{G}$ instead of time integral for I

- example for 2B selfenergy[8]

$$\Sigma^{\gtrless}_{ij}\left(t,t'\right) = \pm\left(\mathrm{i}\hbar\right)^2 \sum_{klpqrs} w_{iklp}\left(t\right) w^{\pm}_{qrjs}\left(t'\right) G^{\gtrless}_{lq}\left(t,t'\right) G^{\gtrless}_{pr}\left(t,t'\right) G^{\lessgtr}_{sk}\left(t',t\right)$$

- respective $\mathcal{G}$ can be identified as

$$\boxed{\mathcal{G}_{ijkl}(t) = \mathrm{i}\hbar \sum_{pqrs} \int_{t_0}^{t} \mathrm{d}\bar{t}\, w^{\pm}_{pqrs}\left(\bar{t}\right) \left[\mathcal{G}^{\mathrm{H},>}_{ijpq}(t,\bar{t})\mathcal{G}^{\mathrm{H},<}_{rskl}(\bar{t},t) - \mathcal{G}^{\mathrm{H},<}_{ijpq}(t,\bar{t})\mathcal{G}^{\mathrm{H},>}_{rskl}(\bar{t},t)\right]}$$

with the two-particle Hartree Green function

$$\mathcal{G}^{\mathrm{H},\gtrless}_{ijkl}(t,t') := G^{\gtrless}_{ik}(t,t')G^{\gtrless}_{jl}(t,t')$$

[8] N. Schlünzen, J.-P. Joost and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020)

- two-particle $\mathcal{G}$ in HF-GKBA

$$\mathcal{G}_{ijkl}(t) = (\mathrm{i}\hbar)^3 \sum_{pqrs} \int_{t_0}^{t} \mathrm{d}\bar{t}\, \mathcal{U}^{(2)}_{ijpq}(t,\bar{t}) \boxed{\Psi^{\pm}_{pqrs}(\bar{t})} \mathcal{U}^{(2)}_{rskl}(\bar{t},t)$$

with the single-time source term (which no longer depends on the outer time)

$$\boxed{\Psi^{\pm}_{ijkl}(t)} = (\mathrm{i}\hbar)^2 \sum_{pqrs} w^{\pm}_{pqrs}(t) \left[\mathcal{G}^{\mathrm{H},>}_{ijpq}(t,t)\mathcal{G}^{\mathrm{H},<}_{rskl}(t,t) - \mathcal{G}^{\mathrm{H},<}_{ijpq}(t,t)\mathcal{G}^{\mathrm{H},>}_{rskl}(t,t) \right]$$

and the two-particle Hartree–Fock time-evolution operators obeying Schrödinger-type EOMs

$$\frac{\mathrm{d}}{\mathrm{d}t}\left[\mathcal{U}^{(2)}_{ijkl}(t,\bar{t})\right] = \frac{1}{\mathrm{i}\hbar} \sum_{pq} h^{(2),\mathrm{HF}}_{ijpq}(t)\, \mathcal{U}^{(2)}_{pqkl}(t,\bar{t})$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left[\mathcal{U}^{(2)}_{ijkl}(\bar{t},t)\right] = -\frac{1}{\mathrm{i}\hbar} \sum_{pq} \mathcal{U}^{(2)}_{ijpq}(\bar{t},t)\, h^{(2),\mathrm{HF}}_{pqkl}(t)$$

with the effective two-particle Hamiltonian

$$h^{(2),\mathrm{HF}}_{ijkl}(t) = \delta_{jl} h^{\mathrm{HF}}_{ik}(t) + \delta_{ik} h^{\mathrm{HF}}_{jl}(t)$$

Time-linear NEGF simulations: the G1–G2 Scheme[9]

C | A | U
Christian-Albrechts-Universität zu Kiel
Mathematisch-Naturwissenschaftliche Fakultät

- full propagation on the time diagonal, as for ordinary HF-GKBA:

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}G^<_{ij}(t) = \left[h^{\mathrm{HF}}, G^<\right]_{ij}(t) + \left[I + I^\dagger\right]_{ij}(t)$$

- but collision integral defined by correlated two-particle Green function

$$I_{ij}(t) = \pm\mathrm{i}\hbar\sum_{klp} w_{iklp}(t)\mathcal{G}_{lpjk}(t)$$

- which obeys an ordinary differential equation

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{G}_{ijkl}(t) = \left[h^{(2),\mathrm{HF}}, \mathcal{G}\right]_{ijkl}(t) + \Psi^{\pm}_{ijkl}(t)$$

$$\Longrightarrow \mathcal{O}(N_\mathrm{t}^1)$$

- the initial values

$$G^{0,<}_{ij} = \pm\frac{1}{\mathrm{i}\hbar}n_{ij}(t_0) =: \pm\frac{1}{\mathrm{i}\hbar}n^0_{ij}\,,$$

$$\mathcal{G}^0_{ijkl} = \frac{1}{(\mathrm{i}\hbar)^2}\left\{n^0_{ijkl} - n^0_{ik}n^0_{jl} \mp n^0_{il}n^0_{jk}\right\}\,,$$

determine the density and the pair correlations existing in the system at the initial time $t = t_0$

[9] N. Schlünzen, J.-P. Joost, and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020)

- other selfenergy approximations can be reformulated in the G1–G2 scheme in similar fashion:[10]

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{G}_{ijkl}(t) = \left[h^{(2),\mathsf{HF}}(t), \mathcal{G}(t)\right]_{ijkl} + \Psi^{\pm}_{ijkl}(t) + \underbrace{L_{ijkl}(t)}_{\text{TPP}} + \underbrace{P_{ijkl}(t)}_{GW} \pm \underbrace{P_{jikl}(t)}_{\text{TPH}}$$

$$L_{ijkl} := \sum_{pq}\left\{\mathfrak{h}^{L}_{ijpq}\mathcal{G}_{pqkl} - \mathcal{G}_{ijpq}\left[\mathfrak{h}^{L}_{klpq}\right]^{*}\right\}, \qquad \mathfrak{h}^{L}_{ijkl} := (\mathrm{i}\hbar)^2\sum_{pq}\left[\mathcal{G}^{\mathsf{H},>}_{ijpq} - \mathcal{G}^{\mathsf{H},<}_{ijpq}\right]w_{pqkl}\,,$$

$$P_{ijkl} := \sum_{pq}\left\{\mathfrak{h}^{\Pi}_{qjpl}\mathcal{G}_{piqk} - \mathcal{G}_{qjpl}\left[\mathfrak{h}^{\Pi}_{qkpi}\right]^{*}\right\}, \qquad \mathfrak{h}^{\Pi}_{ijkl} := \pm(\mathrm{i}\hbar)^2\sum_{pq}w^{\pm}_{qipk}\left[\mathcal{G}^{\mathsf{F},>}_{jplq} - \mathcal{G}^{\mathsf{F},<}_{jplq}\right]$$

and the Hartree/Fock (H/F) two-particle Green functions

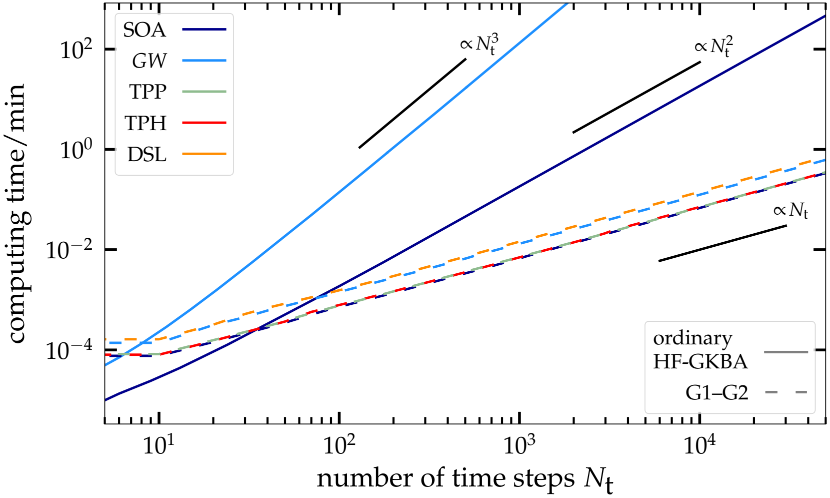
$$\mathcal{G}^{\mathsf{H},\gtrless}_{ijkl}(t) := G^{\gtrless}_{ik}(t,t)G^{\gtrless}_{jl}(t,t)\,, \qquad \mathcal{G}^{\mathsf{F},\gtrless}_{ijkl}(t) := G^{\gtrless}_{il}(t,t)G^{\lessgtr}_{jk}(t,t)$$

- include TPP, GW and TPH terms simultaneously: dynamically-screened-ladder (DSL) approximation. Conserving, applicable to short times. No explicit selfenergy known.[11]
- nonequilibrium generalization of ground state result (Bethe-Salpeter equation)

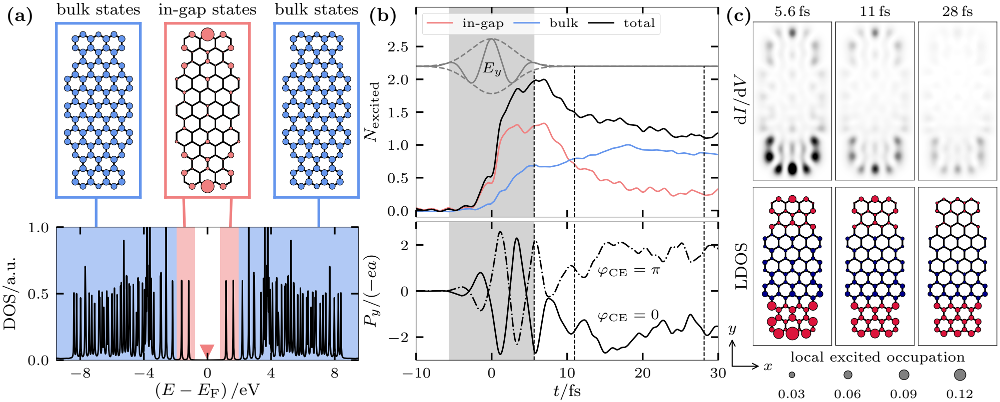
[10] J.-P. Joost, N. Schlünzen, and M. Bonitz, PRB **101**, 245101 (2020), Joost et al., PRB **105**, 165155 (2022);

[11] J.-P. Joost, PhD thesis, Kiel University 2023

- time-linear scaling achieved quickly for all approximations. Example: 10-site Hubbard chain



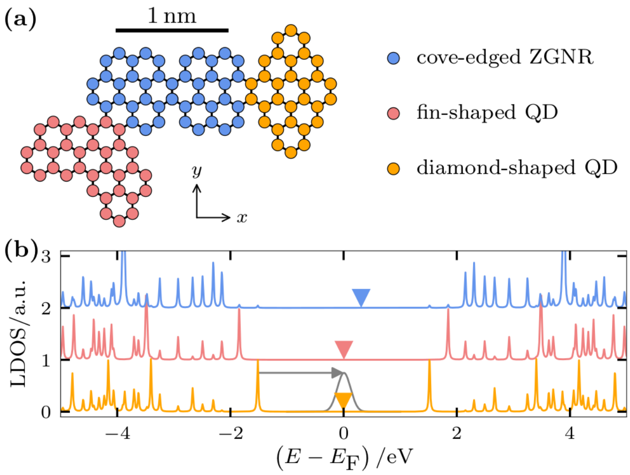
Local Electron Excitation with a spatially uniform laser pulse[12]



- Site-selective excitation on fs time scale. **Promising for PHz nanoelectronics**
- top-bottom symmetry broken by proper choice of carrier-envelope phase
- bulk-edge charge separation persists for 8 fs

[12] J.-P. Joost and M. Bonitz, Phys. Rev. Res. (Lett.) 2025

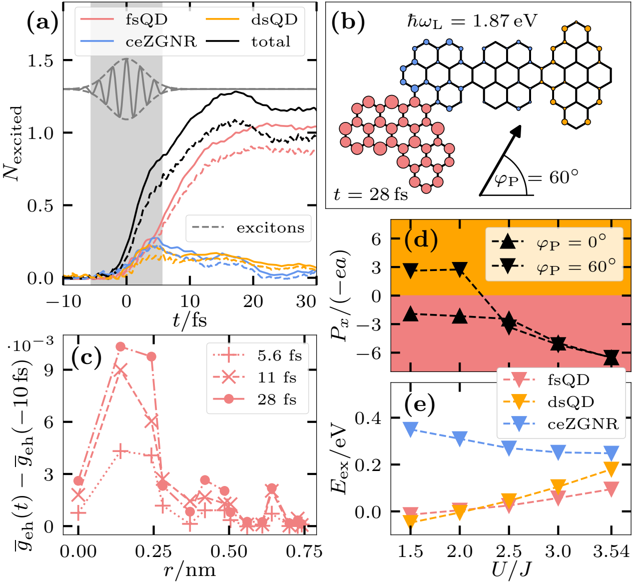
Enhance charge separation: optimized geometry[13]



- small orbital overlap between three system parts
- triangles: exciton binding energies, selective excitation possible

[13] J.-P. Joost and M. Bonitz, Phys. Rev. Res. (Lett.) 2025

Selective excitation of excitons in red QD[14]



[14] J.-P. Joost and M. Bonitz, Phys. Rev. Res. (Lett.) 2025

NEGF-based Quantum Fluctuations Approach (QFA)[15]

- overcome G1-G2 bottleneck: **memory-costly four-point quantities** ($\mathcal{G}_2$)
- Expectation values and fluctuations of t-diagonal Green functions: $\hat{G} = G + \delta\hat{G}$

$$G^{>}_{ij}(t) = \langle \hat{G}^{>}_{ij}(t) \rangle, \qquad \hat{G}^{>}_{ij}(t) = \frac{1}{\mathrm{i}\hbar}\, \hat{a}_i(t)\hat{a}^{\dagger}_j(t)$$

$$G^{<}_{ij}(t) = \langle \hat{G}^{<}_{ij}(t) \rangle, \qquad \hat{G}^{<}_{ij}(t) = \pm\frac{1}{\mathrm{i}\hbar}\, \hat{a}^{\dagger}_j(t)\hat{a}_i(t)$$

$$\delta\hat{G}_{ij}(t) := \delta\hat{G}^{<}_{ij}(t) = \delta\hat{G}^{>}_{ij}(t)$$

- Extension to N-particle fluctuations:

$$L^{(N)}_{i_1 i_2 \dots i_N; j_1 j_2 \dots j_N}(t) := \langle \delta\hat{G}_{i_1 j_1}(t)\delta\hat{G}_{i_2 j_2}(t)\dots\delta\hat{G}_{i_N j_N}(t) \rangle$$

- Short notations: $L_{ijkl}(t) := L^{(2)}_{ij;kl}(t) = G^{(2)}_{ijkl}(t) - G_{ik}(t)G_{jl}(t)\,,$ XC function

- I.) Replace two-particle NEGF $\mathcal{G}_2$ via $L_{ijkl}(t) = \pm G^{>}_{il}(t)G^{<}_{jk}(t) + \mathcal{G}_{ijkl}(t)$

[15] E. Schroedter, J.-P. Joost, and M. Bonitz, (Ukrainian) Cond. Matt. Phys. **25**, 23401 (2022);
classical theory: Yu.L. Klimontovich, cf. E. Schroedter *et al.*, Contrib. Plasma Phys. **64**, e202400015 (2024) and M. Bonitz and A. Zagorodny, Contrib. Plasma Phys. **64**, e202400014 (2024), special issue of CPP
for more details on the quantum fluctuations approach, see publications on our web page

approximation

$$\text{Full NEGF} \xrightarrow{\hspace{4em}} \text{HF-GKBA}$$

$$\mathcal{O}(N_{\mathrm{t}}^3) \qquad\qquad \mathcal{O}(N_{\mathrm{t}}^2)$$

- HF-GKBA recovers Boltzmann-type kinetic equations and overcomes their problems: total energy conservation, correct short-time dynamics, correlated equilibrium state

NEGF simulations with the G1-G2 scheme: Summary and Discussion

$$\text{Full NEGF} \xrightarrow{\textbf{approximation}} \boxed{\text{HF-GKBA} \xrightarrow{\underline{\textbf{no}}\ \textbf{approximation!}} \text{G1–G2}}$$

$$\mathcal{O}(N_\mathrm{t}^3) \qquad \mathcal{O}(N_\mathrm{t}^2) \qquad \mathcal{O}(N_\mathrm{t}^1)$$

- HF-GKBA recovers Boltzmann-type kinetic equations and overcomes their problems: total energy conservation, correct short-time dynamics, correlated equilibrium state
- G1–G2 calculations can be done in **linear time**[16], typical speed-ups: $\times 10^3$–10^6
- **Full nonequilibrium DSL** (combining dyn. screening and strong coupling) simulations possible
- **Memory bottleneck** for $\mathcal{G}_{ijkl}(t) \rightarrow$ mitigated via quantum fluctuations approach[17]

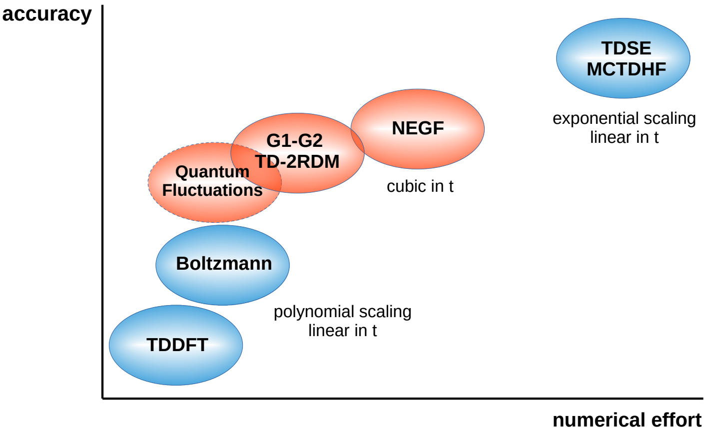
[16] N. Schlünzen, J.-P. Joost and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020);
J.-P. Joost, N. Schlünzen, and M. Bonitz, Phys. Rev. B **101**, 245101 (2020);
J.-P. Joost *et al.*, Phys. Rev. B **105**, 165155 (2022);
Review: M. Bonitz *et al.*, phys. stat. sol. (b), 2300578 (2024)

[17] **Review**: E. Schroedter and M. Bonitz, Contrib. Plasma Phys. **64**:5, e202400015 (2024)

Scaling of selected quantum many-body methods revisited

C | A | U
Christian-Albrechts-Universität zu Kiel
Mathematisch-Naturwissenschaftliche Fakultät

G1–G2 scheme: highly efficient accurate, long and stable correlated quantum dynamics, for systems of any geometry and time scale



External input: parameters of lattice models, efficient atomic basis sets

Outlook[21]

Applications

- highly charged ion impact on 2D quantum materials, fs-neutralization dynamics and secondary electron emission[18], novel diagnostic; plasma-surface interaction
- Laser excitation of macroscopic graphene, TMDCs
- optimized finite graphene clusters for PHz electronics

Method development

- Quantum fluctuations approach for strong coupling[19]
- Spectra and density of states beyond HF and beyond (extended) Koopmans theorem
- extension to open/large systems via embedding selfenergies[20]
- improved selfenergies (3-particle correlations), G1–G2 scheme beyond HF-GKBA

---

[18] Niggas *et al.*, Phys. Rev. Lett. **129**, 086802 (2022)
[19] E. Schroedter, J.-P. Joost, and M. Bonitz, to be published
[20] Balzer *et al.*, PRB **107**, 155141 (2023)
[21] pdf file of talk at https://www.itap.uni-kiel.de/theo-physik/bonitz/talks.html