Nonequilibrium Green functions simulations of femtosecond dynamics of quantum materials

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DESY Hamburg, January 2025

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

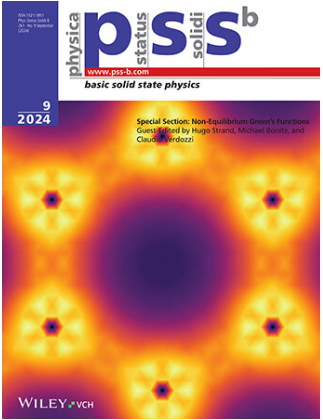


Fig.: G1-G2-simulation of laser excited graphene monolayer (Tim Kalsberger)

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

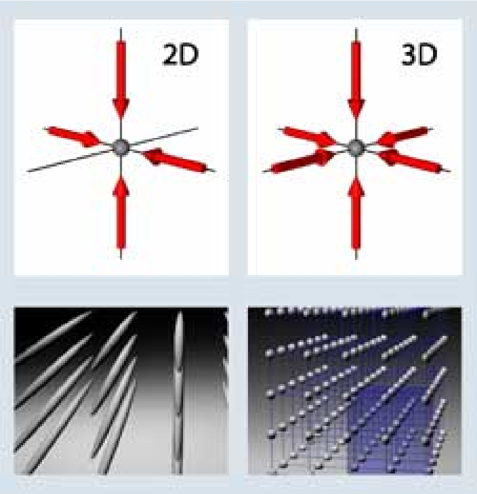
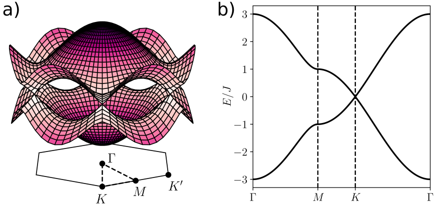


Fig.: M. Greiner (Harvard)

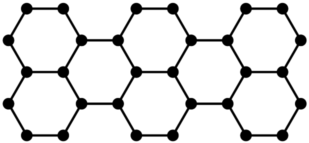
Graphene: high mobility, no bandgap



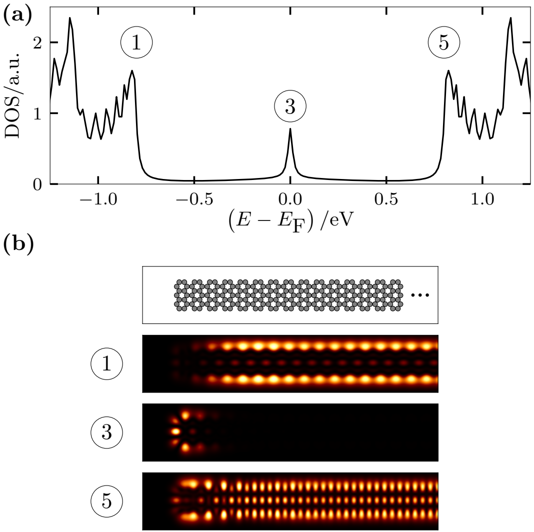
Alternatives : 1. TMDCs:

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)?

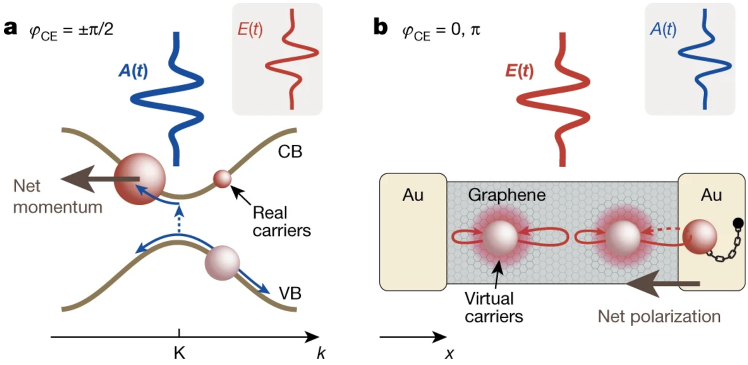
[1] 7 armchair GNR of 504 atoms, GW-NEGF ground state simulation,
J.-P. Joost, A.-P. Jauho, and M. Bonitz, Nano Letters **19**, 9045 (2019)

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 many-electron Hamiltonian

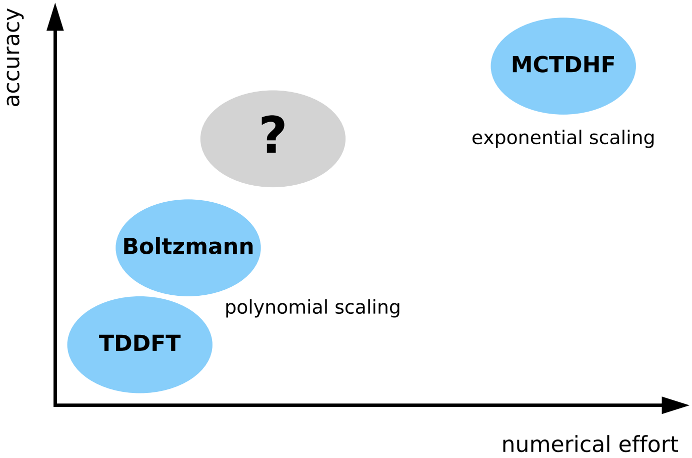
$$H(t) = \underbrace{\sum_{i=1}^{N} h(\boldsymbol{r}_i, t)}_{\text{one-body operators}} + \underbrace{\frac{1}{2}\sum_{i\neq j}^{N} W(\boldsymbol{r}_i, \boldsymbol{r}_j)}_{\textbf{pair-wise}\text{ interactions}}$$

- time-dependent Schrödinger equation (TDSE)

$$i\partial_t \Psi(\boldsymbol{r}_i, \ldots, \boldsymbol{r}_N; t) = H(t)\Psi(\boldsymbol{r}_i, \ldots, \boldsymbol{r}_N; t) \quad \underset{\text{exponential scaling of numerical effort}}{\overset{\text{direct solution}}{\not\Longrightarrow}} \quad \Psi(\boldsymbol{r}_i, \ldots, \boldsymbol{r}_N; t)$$

- **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?

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2nd quantization

- Fock space $\mathcal{F} \ni |n_1, n_2 \ldots\rangle$, $\quad \mathcal{F} = \bigoplus_{N_0 \in \mathbb{N}} \mathcal{F}^{N_0}$, $\quad \mathcal{F}^{N_0} \subset \mathcal{H}^{N_0}$
- $\hat{c}_i, \hat{c}_i^\dagger$ 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 $|\phi_i\rangle$

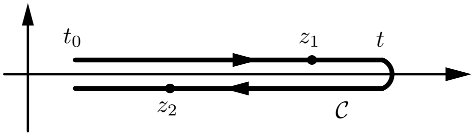
$$\boxed{G_{ij}(z,z') = \frac{\mathrm{i}}{\hbar}\left\langle \hat{T}_{\mathcal{C}} \hat{c}_i(z) \hat{c}_j^\dagger(z') \right\rangle}$$

average with $\hat{\rho}_N$

pure or mixed state

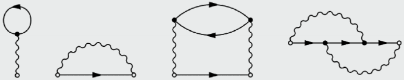
Keldysh–Kadanoff–Baym equations (KBE) on $\mathcal{C}$ (2×2 matrix)[3]:

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



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

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



[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

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

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

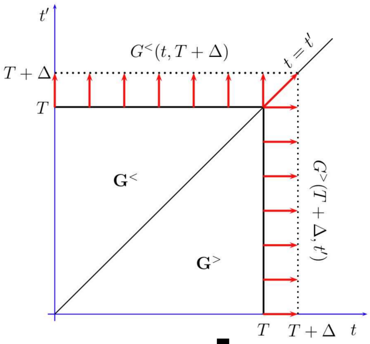
with the effective single-particle **Hartree–Fock Hamiltonian**

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

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

Choice depends on coupling strength, density (filling)

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

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

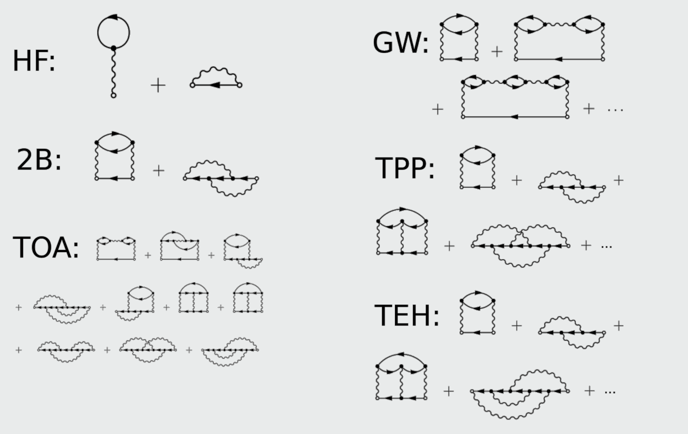
GW: ∞ bubble summation, dynamical screening effects

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): $\sim w^3$

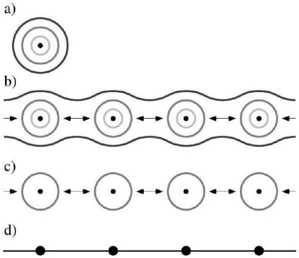
dynamically screened ladder (DSL)*: $\sim \mathrm{2B} + GW + \mathrm{TPP} + \mathrm{TPH}$



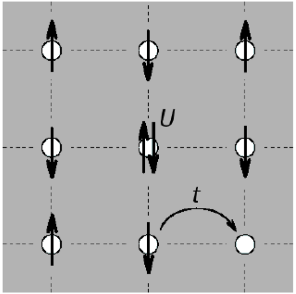
[4]Conserving approximations, nonequilibrium $\Sigma(t,t')$, applies for ultra-short to long times
Review: Schlünzen *et al.*, J. Phys. Cond. Matt. **32**, 103001 (2020); *Joost *et al.*, PRB (2022)

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

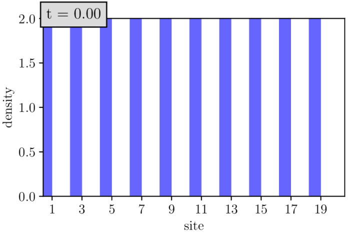
use $J = 1$, on-site repulsion $(U > 0)$ or attraction $(U < 0)$, tunable interaction strength

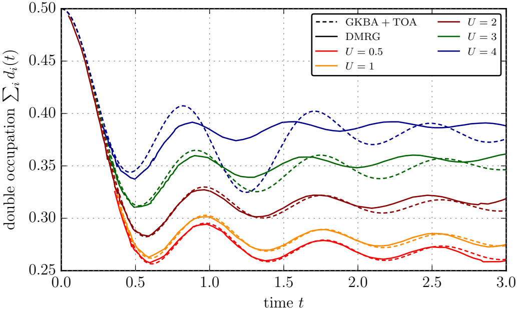
- parameters from electronic structure calculations or experiment
- systematic improvements: extended Hubbard and PPP model [Joost *et al.*, phys. stat. sol. (b) 2019]

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

Initial state:

charge density wave



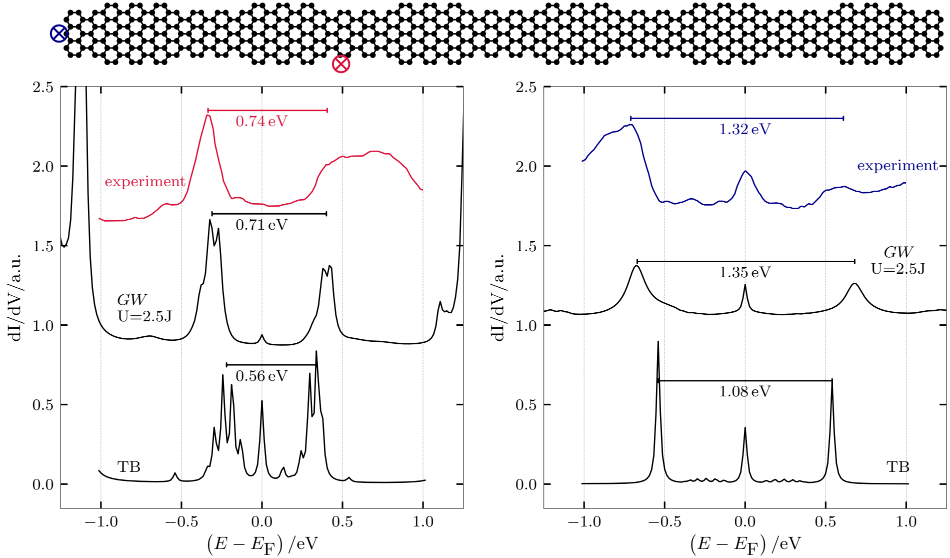


- sensitive observable: total double occupation
- good quality transients NEGF up to $U \simeq$ bandwidth
- accurate long-time behavior of GKBA+T-matrix (not shown)
- performance of different selfenergies vs. coupling and filling[5]

[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]

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

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

- reconstruct off-diagonal NEGF from time diagonal:

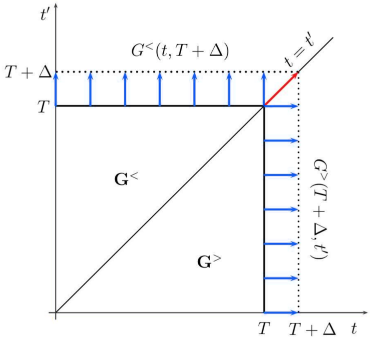
$$G^{\gtrless}_{ij}(t,t') = \pm\left[G^{\mathsf{R}}_{ik}(t,t')\rho^{\gtrless}_{kj}(t') - \rho^{\gtrless}_{ik}(t) G^{\mathsf{A}}_{kj}(t,t')\right]$$

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

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

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

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



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

[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

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

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

time integral off-diagonal functions

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time integral

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

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

time integral 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)

Reformulating the GKBA

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

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

time integral off-diagonal functions **Idea:** solve differential equation for $\mathcal{G}$ instead of time integral for I

- example for 2B selfenergy[8]

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

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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]

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- 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

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

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

- 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)

The G1–G2 Scheme: beyond 2nd Born selfenergy

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

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

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

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

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

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

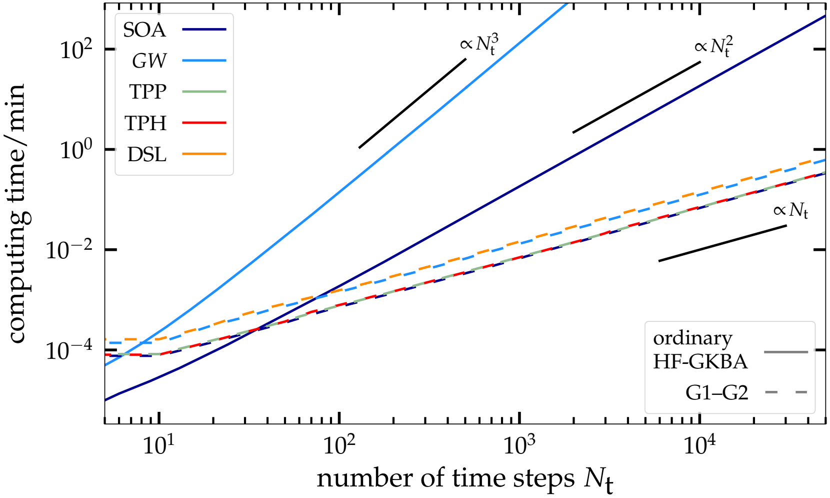
- include TPP, GW and TPH terms simultaneously: dynamically-screened-ladder (DSL) approximation. Conserving, applicable to short times. No explicit selfenergy known.[11]
- nonequilibrium generalization of ground state result (Bethe-Salpeter equation)

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

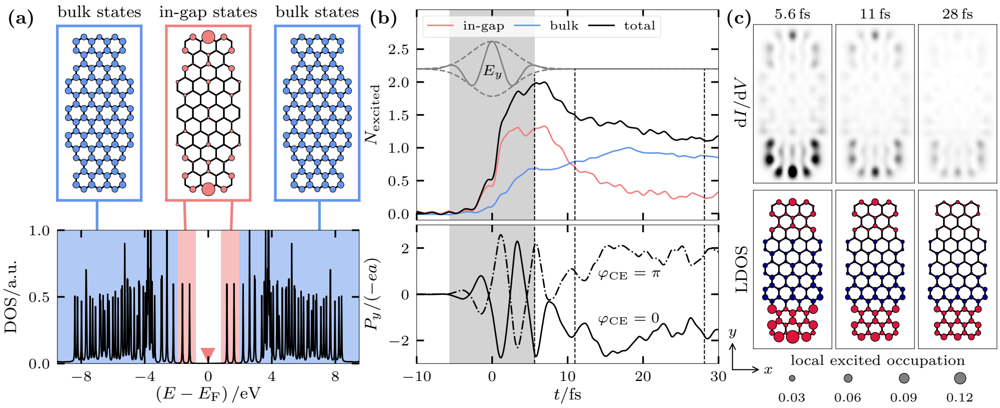
[11] J.-P. Joost, PhD thesis, Kiel University 2023

Numerical Scaling of G1–G2 vs. Standard HF-GKBA

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



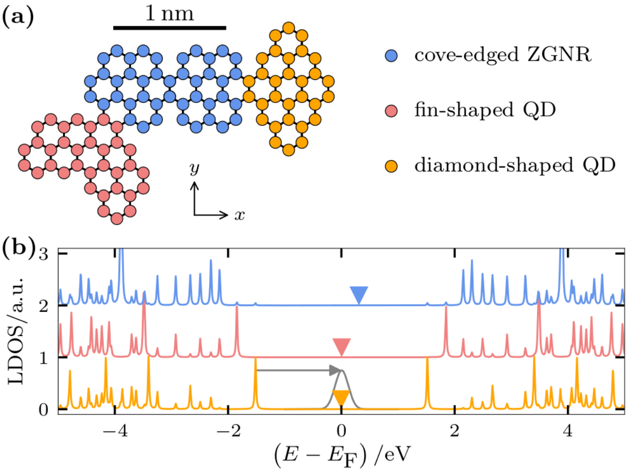
Local Electron Excitation with a spatially uniform laser pulse[12]



- 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, submitted for publication

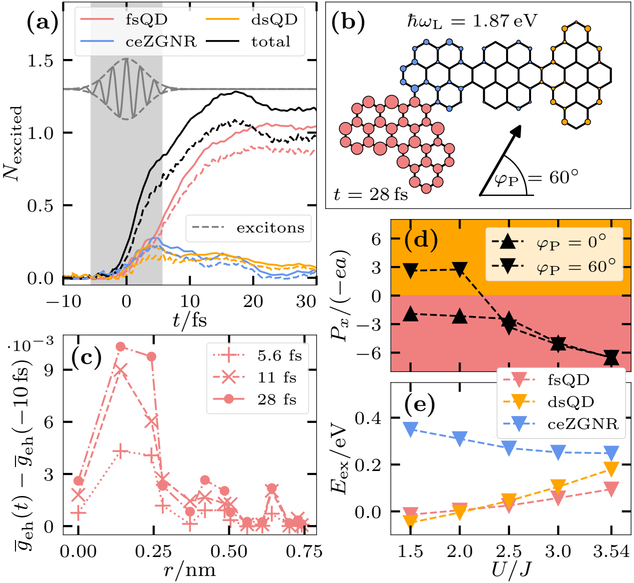
Enhance charge separation: optimized geometry[13]



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

[13] J.-P. Joost and M. Bonitz, submitted for publication

Selective excitation of excitons in red QD[14]



[14] J.-P. Joost and M. Bonitz, submitted for publication

First principles approach to Quantum Fluctuations[15]

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

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

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

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

- Extension to N-particle fluctuations:

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

- Short notations: $L_{ijkl}(t) \coloneqq L^{(2)}_{ij;kl}(t) = G^{(2)}_{ijkl}(t) - G_{ik}(t)G_{jl}(t)\,,$ 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, Cond. Matt. Phys. **25**, 23401 (2022); classical theory. Yu.L. Klimontovich, cf. E. Schroedter *et al.*, Contrib. Plasma Phys. **64**, e202400015 (2024)

In G_1 equation: reformulate collision integral in terms of fluctuations

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

$$h^{\mathrm{H}}_{ij}(t) = h_{ij}(t) + U^{\mathrm{H}}_{ij}(t)\,; \quad U^{\mathrm{H}}_{ij}(t) = \pm i\hbar \sum_{kl} w_{ikjl}(t) G^<_{lk}(t)$$

$$\left[\tilde{I} + \tilde{I}^\dagger\right]_{ij}(t) = \pm i\hbar \sum_{klp} \left\{ w_{iklp}(t) L_{plkj}(t) - w_{kljp}(t) L_{ipkl}(t) \right\}$$

II.) Eliminate dynamics of $L_{ijkl}(t) = \langle \delta\hat{G}_{ik}(t) \cdot \delta\hat{G}_{jl}(t) \rangle$ by propagating $\delta\hat{G}(t)$:

Exact equation for single-particle fluctuation (two-point function)

$$i\hbar\partial_t \delta\hat{G}_{ij}(t) = \left[h^{\mathrm{H}}, \delta\hat{G}\right]_{ij}(t) + \left[\delta\hat{U}^{\mathrm{H}}, G^<\right]_{ij}(t) + \left[\delta\hat{U}^{\mathrm{H}}, \delta\hat{G}\right]_{ij}(t) - \left[\tilde{I} + \tilde{I}^\dagger\right]_{ij}(t)$$

$$\delta\hat{U}^{\mathrm{H}}_{ij}(t) = \pm i\hbar \sum_{kl} w_{ikjl}(t) \delta\hat{G}^<_{lk}(t)$$

[16] E. Schroedter, J.-P. Joost, and M. Bonitz, Cond. Matt. Phys. **25**, 23401 (2022);
E. Schroedter et al., PRB **108**, 205109 (2023)

Stochastic Mean field idea:[17]

- III.) Replace quantum expectation value by semiclassical mean over realizations A^λ, $\langle \hat{A} \rangle \longrightarrow \overline{A^\lambda}$, exactly preserve first two moments
- Random sampling of initial conditions of non-interacting system:

$$\begin{aligned}\overline{\Delta G_{ij}^\lambda(t_0)} &= 0, \\ \overline{\Delta G_{ik}^\lambda(t_0) \Delta G_{jl}^\lambda(t_0)} &= -\frac{1}{2\hbar^2} \delta_{il} \delta_{jk} \{ n_i(1 \pm n_j) + n_j(1 \pm n_i) \}.\end{aligned}$$

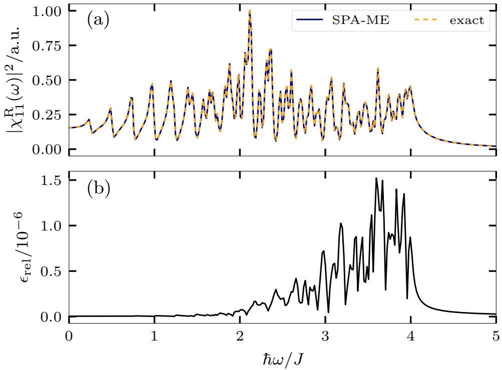
- interactions turned on via adiabatic switching
- Careful test of probability distribution and sampling methods[18]
- IV.) find approximations that have product form: $\mathcal{G}^{\mathrm{app}} \rightarrow L^{\mathrm{app}}(t) = \langle \delta \hat{G}(t) \cdot \delta \hat{G}(t) \rangle$

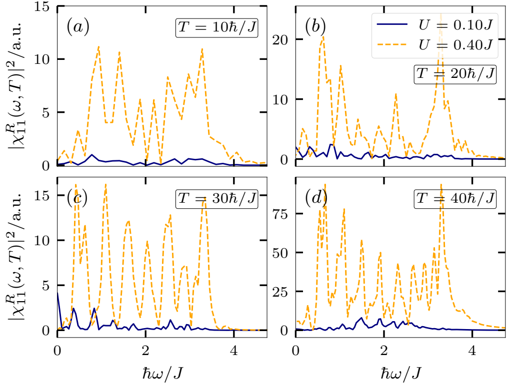
[17] S. Ayik, Phys. Lett. B **658**, 174 (2008);
D. Lacroix, S. Hermanns, C. M. Hinz, and M. Bonitz, PRB **90**, 125112 (2014)

[18] E. Schroedter, J.-P. Joost, and M. Bonitz, Cond. Matt. Phys. **25**, 23401 (2022);
E. Schroedter *et al.*, PRB **108**, 205109 (2023)

The stochastic GW-approximation[19]

Two-time density response function: $\chi^{\mathrm{R}}_{ij}(t,t') = 2\hbar\Theta(t-t')\mathrm{Im}\left[\overline{\Delta G^{\lambda}_{ii}(t)(\Delta G^{\lambda}_{jj}(t'))^{*}}\right]$





Left: Ground state of a half-filled 50 site ring for $U = 0$. **Right:** Nonequilibrium density response, for different times T, after a confinement quench, for a half-filled 30 site ring.

[19] E. Schroedter, B.J. Wurst, J.-P. Joost, and M. Bonitz, Phys. Rev. B **108**, 205109 (2023); equivalent to BSE in GW with HF-GKBA: E. Schroedter and M. Bonitz, phys. stat. sol. (b), 2300564 (2024)

$$\text{Full NEGF} \xrightarrow{\textbf{approximation}} \text{HF-GKBA}$$

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

- 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

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

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

- 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**[20], 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[21]

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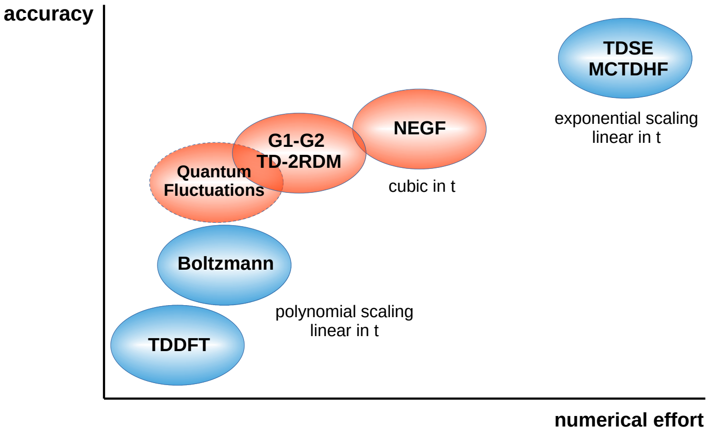
[20] N. Schlünzen, J.-P. Joost and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020);
J.-P. Joost, N. Schlünzen, and M. Bonitz, Phys. Rev. B **101**, 245101 (2020);
J.-P. Joost *et al.*, Phys. Rev. B **105**, 165155 (2022);
Review: M. Bonitz *et al.*, phys. stat. sol. (b), 2300578 (2024)

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

Scaling of selected quantum many-body methods revisited

CAU
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[25]

Applications

- highly charged ion impact on 2D quantum materials, fs-neutralization dynamics and secondary electron emission[22], 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[23]
- Spectra and density of states beyond HF and beyond (extended) Koopmans theorem
- extension to open/large systems via embedding selfenergies[24]
- improved selfenergies (3-particle correlations), G1–G2 scheme beyond HF-GKBA

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