Quantum Dynamics of finite Hubbard clusters

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Missing: David, Lukas, Torben, Hanno, Patrick

DFG

Bundesministerium
für Bildung
und Forschung

Outline

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- **High-intensity lasers, free electron lasers**
  - strong nonlinear excitation of matter
  - high photon energy: core level excitation
  - localized excitation: spatial inhomogeneity
- **Ultra-short pulses**
  - (sub-)fs dynamics of atoms, molecules, solids
  - sub-fs dynamics of electronic correlations
- **Need: Nonequilibrium many-body theory**
  - conservation laws on all time scales
  - linear and nonlinear response
  - macroscopic to finite (inhomogeneous) systems

Theoretical approaches to finite correlated systems in nonequilibrium

I. Wave function based methods (pure state)
   - Solution of Schrödinger equation, Full CI
   - Multiconfiguration time-dependent Hartree-Fock (MCTDHF, [1])
   - Restricted active space CI (TDRAS-CI, [1])

   $\Rightarrow$ talk by Christopher Hinz

II. Statistical approaches (mixed ensemble)
   - Nonequilibrium Green functions (NEGF, 2-time fcts [2])
   - Reduced density operator techniques (1-time fcts [3])
   - NEGF with generalized KB ansatz (GKBA)

[1] D. Hochstuhl, C. Hinz, and M. Bonitz, EPJ-ST (2013), arXiv: 1310.xxxx
[2] K. Balzer, and M. Bonitz, Springer Lecture Notes in Physics **867** (2013)
[3] M. Bonitz, *Quantum Kinetic Theory*, Teubner 1998

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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: $\hat{H}(t) = \underbrace{\sum_{k,m} h^0_{km}\hat{c}^\dagger_k\hat{c}_m}_{\hat{H}_0} + \underbrace{\frac{1}{2}\sum_{k,l,m,n} w_{klmn}\,\hat{c}^\dagger_k\hat{c}^\dagger_m\hat{c}_n\hat{c}_l}_{\hat{W}} + F(t)$

Particle interaction w_{klmn}

- Only electron dynamics
- Coulomb interaction

Time-dependent excitation F(t)

- Single-particle type
- Optical/Laser-induced

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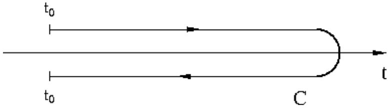
time-ordered one-particle Nonequilibrium Green function,
two times $z, z' \in \mathcal{C}$ ("Keldysh contour"), arbitrary one-particle basis $|\phi_i\rangle$

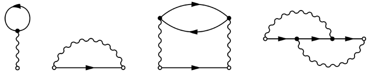
$$G^{(1)}_{ij}(z, z') = \frac{\mathrm{i}}{\hbar} \left\langle \hat{T}_{\mathcal{C}} \hat{c}_i(z) \hat{c}^\dagger_j(z') \right\rangle$$

Keldysh–Kadanoff–Baym equation (KBE) on $\mathcal{C}$:

$$\sum_k \left\{ \mathrm{i}\hbar \frac{\partial}{\partial z} \delta_{ik} - h_{ik}(z) \right\} G^{(1)}_{kj}(z, z') = \delta_{\mathcal{C}}(z, z')\delta_{ij} - \mathrm{i}\hbar \sum_{klm} \int_{\mathcal{C}} \mathrm{d}\bar{z}\, w_{iklm}(z^+, \bar{z}) G^{(2)}_{lmjk}(z, \bar{z}; z', \bar{z}^+)$$



- $\int_{\mathcal{C}} w G^{(2)} \rightarrow \int_{\mathcal{C}} \Sigma G^{(1)}$, Selfenergy
- Nonequilibrium Diagram technique
  Example: Hartree-Fock + Second Born selfenergy



KBE: first equation of Martin–Schwinger hierarchy for $G^{(1)}, G^{(2)} \ldots G^{(n)}$

- Contour Green function mapped to real-time matrix Green function

$$\mathbf{G_{ij}} = \begin{pmatrix} G^{\mathsf{R}}_{ij} & G^{<}_{ij} \\ 0 & G^{\mathsf{A}}_{ij} \end{pmatrix}$$

$$G^{<}_{ij}(t_1, t_2) = \mp \mathrm{i} \left\langle \hat{c}^{\dagger}_j(t_2) \hat{c}_i(t_1) \right\rangle$$

$$G^{>}_{ij}(t_1, t_2) = -\mathrm{i} \left\langle \hat{c}_i(t_1) \hat{c}^{\dagger}_j(t_2) \right\rangle$$

- Propagators

$$G^{\mathsf{R/A}}(t_1, t_2) = \pm\theta \left[\pm(t_1 - t_2)\right] \left\{ G^{>}(t_1, t_2) - G^{<}(t_1, t_2) \right\}$$

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

$$\left[\mathrm{i}\partial_{t_1} - h_0(t_1)\right] G^{<}(t_1, t_2) = \int \mathrm{d}t_3\, \Sigma^{\mathsf{R}}(t_1, t_3) G^{<}(t_3, t_2) + \int \mathrm{d}t_3\, \Sigma^{<}(t_1, t_3) G^{\mathsf{A}}(t_3, t_2)\,,$$

$$G^{<}(t_1, t_2) \left[-\mathrm{i}\partial_{t_2} - h_0(t_2)\right] = \int \mathrm{d}t_3\, G^{\mathsf{R}}(t_1, t_3) \Sigma^{<}(t_3, t_2) + \int \mathrm{d}t_3\, \Sigma^{\mathsf{A}}(t_1, t_3) G^{<}(t_3, t_2)$$

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Time-dependent single-particle operator expectation value

$$\langle \hat{O} \rangle(t) = \mp\, i \int dx \left[o(x' t)\, g^{<}(xt, x' t) \right]_{x=x'}$$

- Particle density

$$\langle \hat{n}(x,t) \rangle = n(1) = \mp\, i\, g^{<}(1,1)$$

- Density matrix

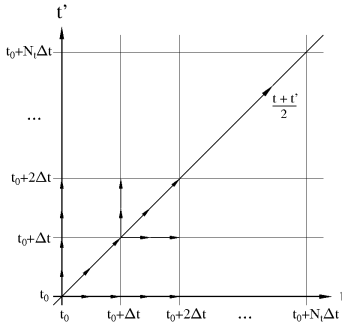
$$\rho(x_1, x_1', t) = \mp\, i\, g^{<}(1,1')\Big|_{t_1 = t_1'}$$

- Current density: $\langle \hat{j}(1) \rangle = \mp\, i \left[\left(\frac{\nabla_1}{2i} - \frac{\nabla_{1'}}{2i} + A(1) \right)\, g^{<}(1,1') \right]_{1'=1}$

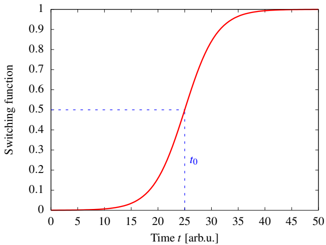
Interaction energy (two-particle observable, [Baym/Kadanoff])

$$\langle \hat{V}_{12} \rangle(t) = \pm\, i\, \frac{\mathcal{V}}{4} \int \frac{d\vec{p}}{(2\pi\hbar)^3} \left\{ (i\,\partial_t - i\,\partial_{t'}) - \frac{p^2}{m} \right\} g^{<}(\vec{p}, t, t')|_{t=t'}$$

Full two-time solutions: Danielewicz, Schäfer, Köhler/Kwong, Bonitz/Semkat, Haug, Jahnke, van Leeuwen, Stefanucci, Verdozzi, Berges, Garny ...



2. adiabatically slow switch-on of interaction for $t, t' \leq t_0$ [1, 2]

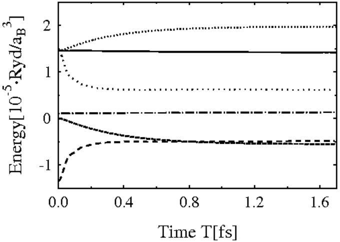


1. Uncorrelated initial state

3. solve KBE in $t - t'$ plane for $g^{\gtrless}(t, t')$

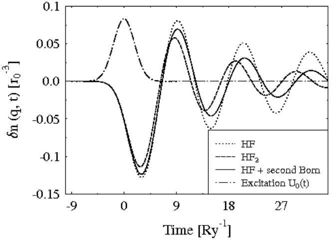
[1] A. Rios et al., Ann. Phys. **326**, 1274 (2011), [2] S. Hermanns et al., Phys. Scr. **T151**, 014036 (2012)

1. perfect conservation of total energy
2. accurate short-time dynamics:
   phase 1: correlation dynamics
   2: relaxation of $f(p)$, occupations



Example: electrons in dense hydrogen, interaction quench [1]

3. accurate long-time behavior: spectral functions and high-order correlated spectra from real-time KBE dynamics (via Fourier transform) [2]



4. extended to optical absorption, double excitations [3] etc.

[1] MB and D. Semkat, *Introduction to Computational Methods in Many-Body Physics*, Rinton Press 2006,

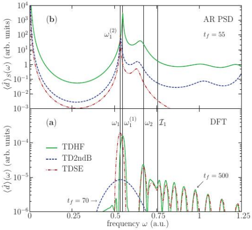
[2] N. Kwong and MB, PRL **84**, 1768 (2000), [3] K. Balzer, S. Hermanns, MB, EPL **98**, 67002 (2012)

- few-electron atoms, molecules [PRA **81**, 022510 (2010), PRA **82**, 033427 (2010)]

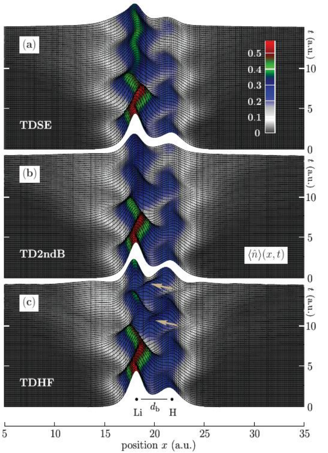
1D He ground state

n_g (n_b)	Number of τ-grid points	E^{HF}_{gs} [a.u.]
Hartree-Fock		
	4 (43)	−2.22
	9 (98)	−2.224209
	14 (153)	−2.2242096
Second Born		E^{2ndB}_{gs} [a.u.]
14 (153)	101	−2.23
14 (153)	301	−2.2334
14 (153)	601	−2.23341
14 (153)	1001	−2.233419
TDSE (exact)		E^{TDSE}_{gs} [a.u.]
		−2.2382578

1D He dipole spectra



LiH XUV-pulse excitation



* Pioneered by N.E. Dahlen and R. van Leeuwen

Numerical challenges of NEGF calculations

- Complicated structure of interaction w_{klmn} and selfenergy Σ
- Collision intergrals involve integrations over whole past
- CPU time $\sim N_t^3$, RAM $\sim N_t^2$

Typical computational parameters

- Spatial basis size: $N_b = 70$
- Time steps: $N_t = 10000$
- RAM consumption: 2 TB
- number of CPUs used: 2048
- total computation time: 2-3 days

Solutions[1]

- Finite-Element Discrete Variable Representation [PRA **81**, 022510 (2010)]
- Generalized Kadanoff–Baym ansatz [Phys. Scr. **T151**, 014036 ('12), JPCS **427**, 012006 ('13)]
- Adiabatic switch-on of interaction [Phys. Scr. **T151**, 014036 ('12)]
- Parallelization [PRA **82**, 033427 (2010)] and GPU computing

[1] K. Balzer, M. Bonitz, Lecture Notes in Phys. vol. 867 (2013)

The Generalized Kadanoff-Baym Ansatz

Equivalent form of the KBE [*Lipavskii et. al.*]:

- For times $t_1 > t_2 > t_0$:

$$G^<(t_1,t_2) = -G^{\mathsf{R}}(t_1,t_2)\rho(t_2) + \int_{t_2}^{t_1} \mathrm{d}t_3 \int_{t_0}^{t_2} \mathrm{d}t_4\, G^{\mathsf{R}}(t_1,t_3)\Sigma^<(t_3,t_4)G^{\mathsf{A}}(t_4,t_2) + \int_{t_2}^{t_1} \mathrm{d}t_3 \int_{t_0}^{t_2} \mathrm{d}t_4\, G^{\mathsf{R}}(t_1,t_3)\Sigma^{\mathsf{R}}(t_3,t_4)G^<(t_4,t_2)\,.$$

- For times $t_0 < t_1 < t_2$:

$$G^<(t_1,t_2) = \rho(t_1)G^{\mathsf{A}}(t_1,t_2) + \int_{t_2}^{t_1} \mathrm{d}t_3 \int_{t_0}^{t_2} \mathrm{d}t_4\, G^{\mathsf{R}}(t_1,t_3)\Sigma^<(t_3,t_4)G^{\mathsf{A}}(t_4,t_2) + \int_{t_2}^{t_1} \mathrm{d}t_3 \int_{t_0}^{t_2} \mathrm{d}t_3\, G^<(t,t_3)\Sigma^{\mathsf{A}}(t_3,t_4)G^{\mathsf{A}}(t_4,t_2)\,.$$

The generalized Kadanoff-Baym ansatz II

- Idea of the GKBA: lowest order solution

$$\boxed{G^{\gtrless}_{\mathsf{GKBA}}(t_1, t_2) = -G^{\mathsf{R}}(t_1, t_2) f^{\gtrless}(t_2) + f^{\gtrless}(t_1) G^{\mathsf{A}}(t_1, t_2)}$$ [1]

$$f^{<}(t) = f(t) = \pm \mathrm{i} G^{<}(t, t), \quad f^{>}(t) = 1 \pm f^{<}(t)$$

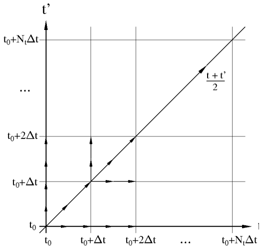
- correct causal structure, non-Markovian, no near-equilibrium assumption[2],
- Reduction to single-time quantities by use of HF propagators

$$G^{\mathsf{R/A}}_{\mathsf{HF}}(t_1, t_2) = \mp \mathrm{i}\theta[\pm(t_1 - t_2)] \exp\left(-\mathrm{i}\int_{t_2}^{t_1} \mathrm{d}t_3\, h_{\mathsf{HF}}(t_3)\right)$$

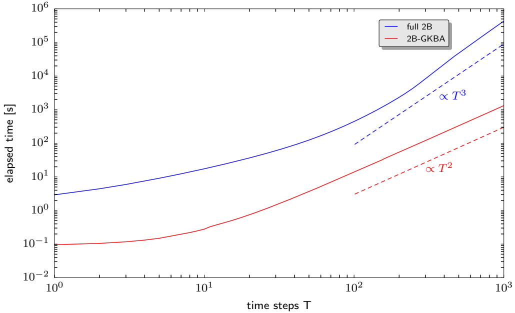
- HF-GKBA: same conservation properties as two-time approximation
- damped propagators, local approximation violate E-conservation [3]

[1] P. Lipavsky, V. Spicka and B. Velicky Phys. Rev. B **34**, 6933 (1986), [2] M. Bonitz, *Quantum Kinetic Theory*
[3] M. Bonitz, D. Semkat, H. Haug, Eur. Phys. J. B **9**, 309 (1999)

Peformance gain with the GKBA



time stepping along diagonal only. Full memory retained.



S. Hermanns, K. Balzer, and M. Bonitz, *Phys. Scripta* **T151**, 014036 (2012)

we use about $5 \cdot 10^3 \ldots 5 \cdot 10^4$ time steps for the adiabatic switching and $10^5 \ldots 10^6$ for the excitation and relaxation.

Summary 1: Real-time NEGF, GKBA

Generalized non-Markovian quantum kinetic equations

- pure and mixed state description
- total energy conserving, nonequilibrium diagram technique
- correct correlated asymptotic state, spectra
- valid for arbitrary fast processes

Direct access to nonlinear and short-time physics

1. strong spatially inhomogeneous laser excitation feasible (plan)
2. non-trivial short-time dynamics, interaction quenches:
   correlation build up[2], prethermalization[3]— universal behavior?

[2] MB *et al.*, J. Phys. Cond. Matt. **8**, 6057 (1996); PRE **56**, 1246 (1997)
[3] Berges *et al.*, PRL **93**, 14303 (2004); Kehrein *et al.*, NJP **12**, 055016 (2010)

Outline

BBGKY-hierarchy (Bogolyubov, Born, Green, Kirkwood, Yvon)

1. N-particle density operator:

$$\rho_N = \sum_k W_k \, |\Psi^{(k)}_{1\ldots N}\rangle \langle \Psi^{(k)}_{1\ldots N}| , \quad \sum_k W_k = 1, \quad \mathrm{Tr}_{1\ldots N}\rho_N = 1$$

$$i\hbar \frac{\partial}{\partial t}\rho_N - \Big[H_{1\ldots N} \, , \, \rho_N \Big]_-(t) = 0 \, , \quad \text{von Neumann eqn.}$$

2. reduced density operators:

$$F_{1\ldots s} = \frac{N!}{(N-s)!} \mathrm{Tr}_{s+1\ldots N}\rho_N \, , \quad \mathrm{Tr}_{1\ldots s} F_{1\ldots s} = \frac{N!}{(N-s)!} \, ,$$

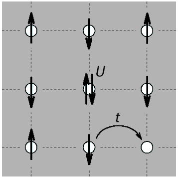
3. partial trace of von Neumann eqn. $\Rightarrow$ BBGKY-hierarchy

M. Bonitz, *Quantum Kinetic Theory*, Teubner 1998
Wang S.-J., W. Cassing, Ann. Phys. **159**, 328 (1985)
A. Akbari et al., Phys. Rev. B **85**, 235121 (2012)

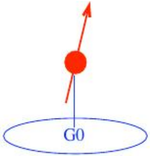
Outline

Lattice models

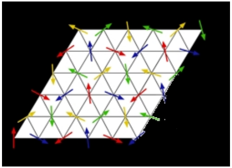
Hubbard



Anderson impurity



Heisenberg

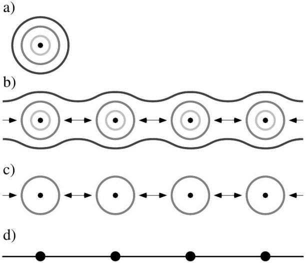


- simplification of the many-body problem
  - localized sites
  - interaction and exchange effects tractable
- macroscopic and finite systems

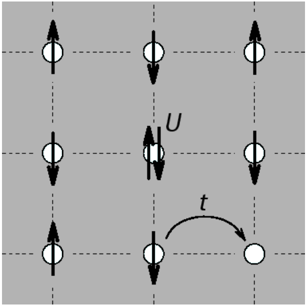
- derived from many-body theory for many systems
  - condensed matter (transition metal oxides, . . .)
  - ultracold particles in optical lattices
  - molecules

The Hubbard model

- Simple, but versatile model for solid state systems
- Suitable for single band, small bandwidth



$\Rightarrow$



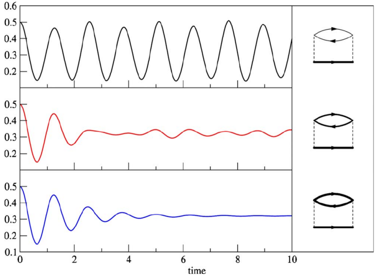
$$\hat{H}(t) = J \sum_{ij,\,\alpha} h_{ij}\, \hat{c}^{\dagger}_{i\alpha} \hat{c}_{j\alpha} + U \sum_i \hat{c}^{\dagger}_{i\uparrow} \hat{c}_{i\uparrow} \hat{c}^{\dagger}_{i\downarrow} \hat{c}_{i\downarrow} + \sum_{ij,\alpha\beta} f_{ij,\alpha\beta}(t)\, \hat{c}^{\dagger}_{i\alpha} \hat{c}_{j\beta}$$

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

Two-site Hubbard—strong excitation

Problems of NEGF in second Born: $N = 2, n = 1/2, U = 1$ [1],
Excitation matrix: $f_{ij,\alpha\beta}(t) = w_0 \delta_{i,1}\delta_{j,1}\delta_{\alpha,\beta}\Theta(t)\,, \quad w_0 = 5.0 J^{-1}$

- time-dependent density, KBE for various degrees of selfconsistency [1]
  artif. damping, mult. steady states



[1] P. von Friesen, C. Verdozzi, and C.O. Almbladh, Phys. Rev. B **82**, 155108 (2010)

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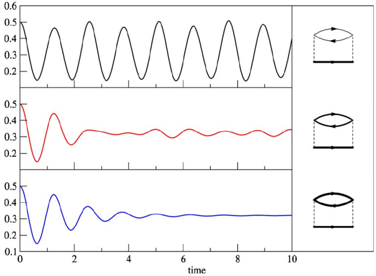
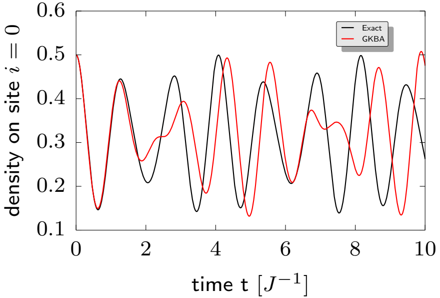
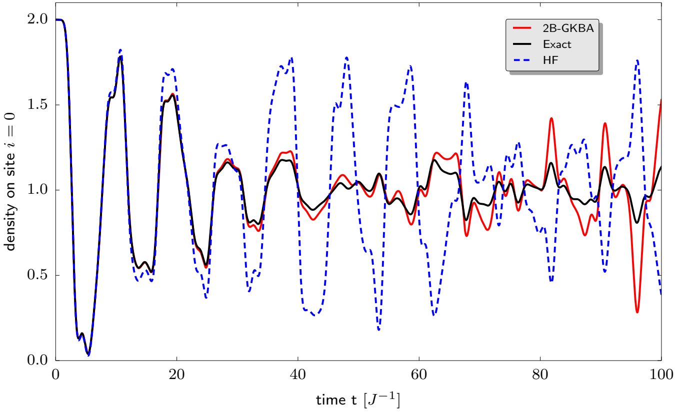


- GKBA: no damping
  selfconsistency problem cured [2]



[1] P. von Friesen, C. Verdozzi, and C.O. Almbladh, Phys. Rev. B **82**, 155108 (2010)
[2] S. Hermanns, and M. Bonitz, Phys. Rev. B (2013)

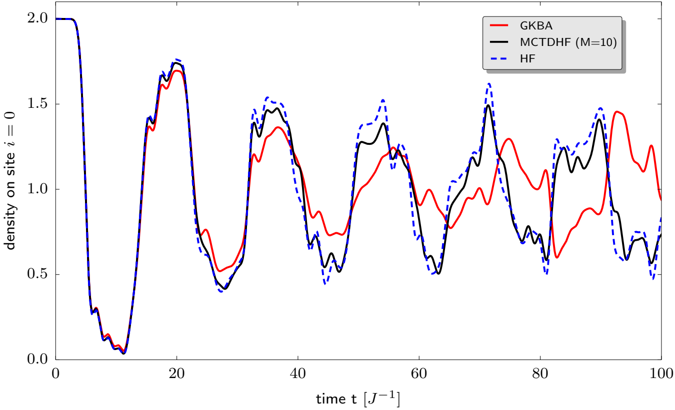
Sites $0 - 3$ doubly occupied, $4 - 7$ empty



failure of HF, good performance of GKBA up to longer times ($t \sim 50$)
GKBA improves with particle number

Half filling—noneq. initial state $N = 16, \ U = 0.1$

Sites $0 - 7$ doubly occupied, $8 - 15$ empty



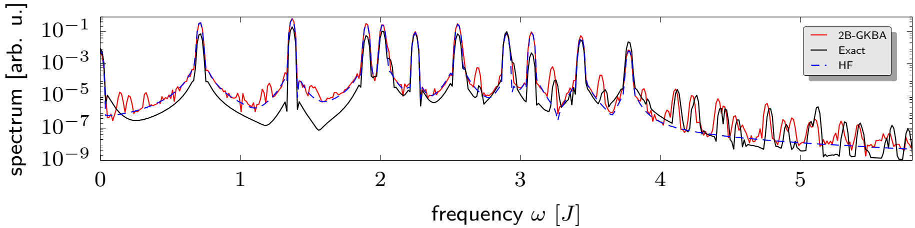
no FCI data, failure of HF (and MCTDHF), expect predictive capability of GKBA

Spectrum—from long real-time propagation

- GKBA with 2ndBorn selfenergy scales as $\mathcal{O}(T^2)$
- 1-3 orders of magnitude longer propagation compared to two-time KBE
- Increased resolution of spectra. Capture double excitations

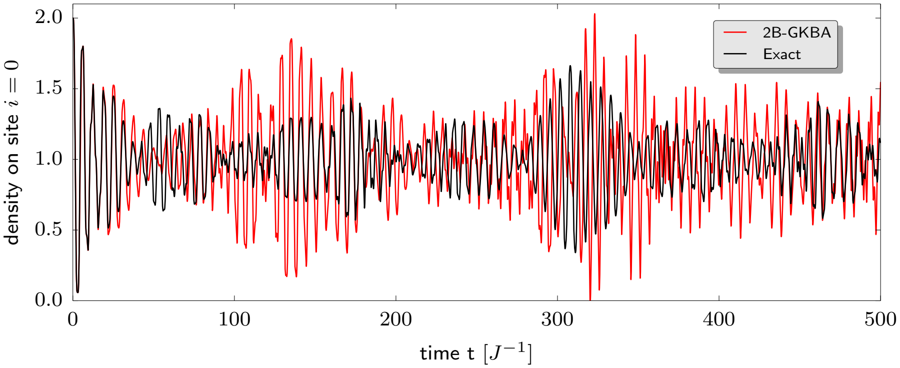
Real-time propagation following weak excitation and Fourier transform

Example: $N = 8, n = 1/2, U = 0.1$



Long relaxation
exact result vs. GKBA, $N = 4,\ n = 1/2,\ U = 0.1$

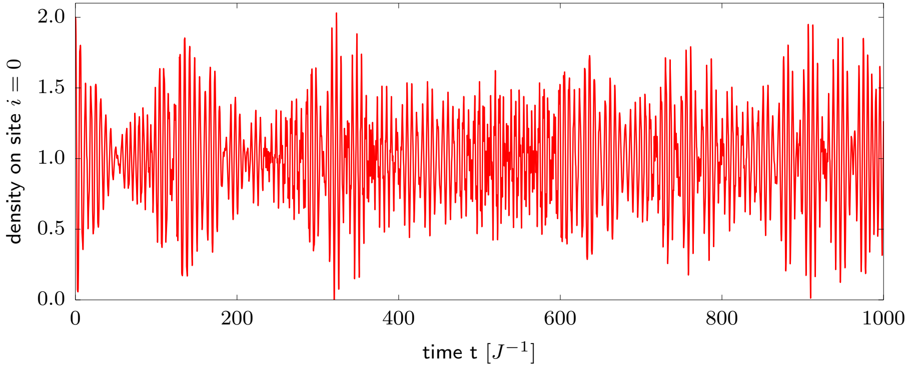
Sites $0 - 1$ doubly occupied, $2 - 3$ empty



GKBA: long-time stability, no divergencies
qualitatively correct up to $t \sim 180$

Increase relaxation duration by 2 (GKBA)
$N = 4,\ n = 1/2,\ U = 0.1$

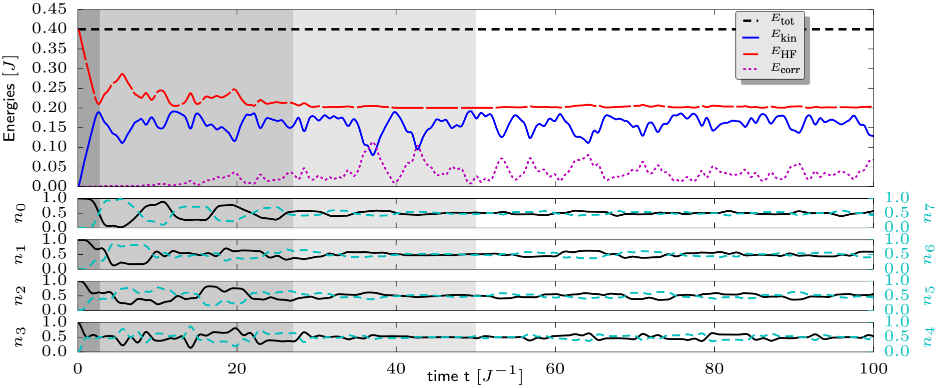
Sites $0 - 1$ doubly occupied, $2 - 3$ empty



GKBA: long-time stability confirmed

Short-time dynamics: four stages
exact calculation, $N = 8,\ n = 1/2,\ U = 0.1$

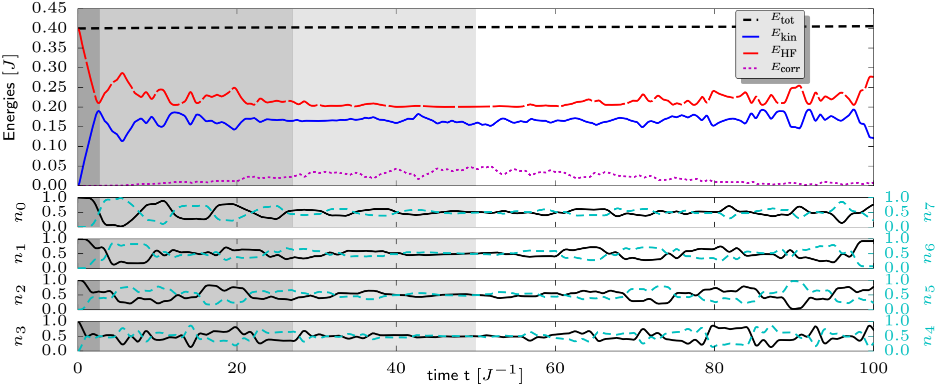
Sites $0 - 3$ doubly occupied, $4 - 7$ empty



I: $t \leq 3$, ballistic expansion (feature of inhomogeneity)
II: $t \leq 25$, correlation build-up/saturation of HF energy
III: $t \leq 50$, one-particle equilibration (occupations)
IV: $t \geq 50$, weak revivals of occupations

Short-time dynamics with GKBA
$N = 8,\ n = 1/2,\ U = 0.1$

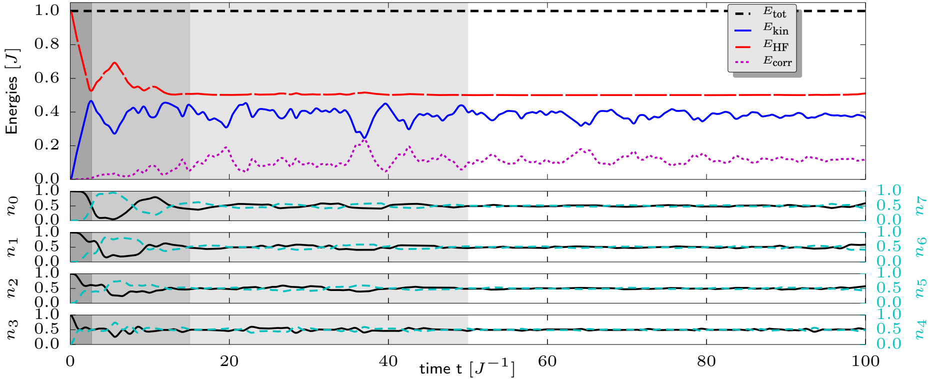
Sites $0 - 3$ doubly occupied, $4 - 7$ empty



GKBA: correctly describes stages I-III, (weaker dynamics of $E_{\text{kin}}, E_{\text{corr}}$)
incorrect: exaggerated revivals (stage IV)

Short-time dynamics ($U = 0.25$): four stages exact calculation: $N = 8$, $n = 1/2$

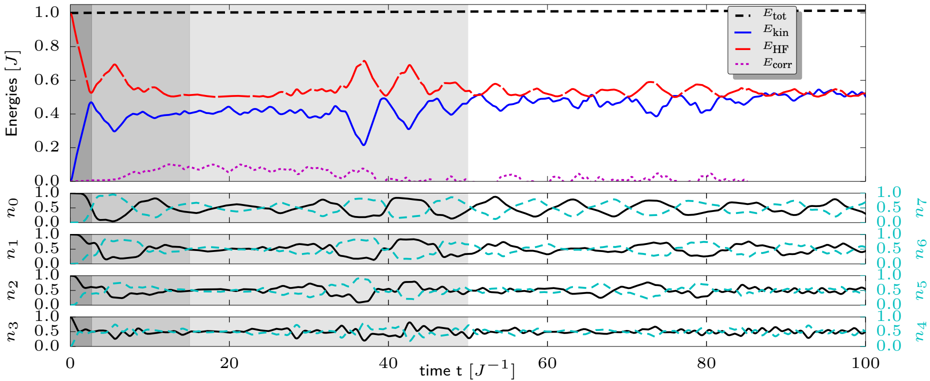
Sites $0 - 3$ doubly occupied, $4 - 7$ empty



I: $t \leq 3$, ballistic expansion (feature of inhomogeneity)
II: $t \leq 15$, correlation build-up/saturation of HF energy
III: $t \leq 50$, one-particle equilibration (occupations)
IV: $t \geq 50$, very weak revivals of occupations

Short-time dynamics with GKBA
$N = 8,\ n = 1/2,\ U = 0.25$

Sites $0-3$ doubly occupied, $4-7$ empty



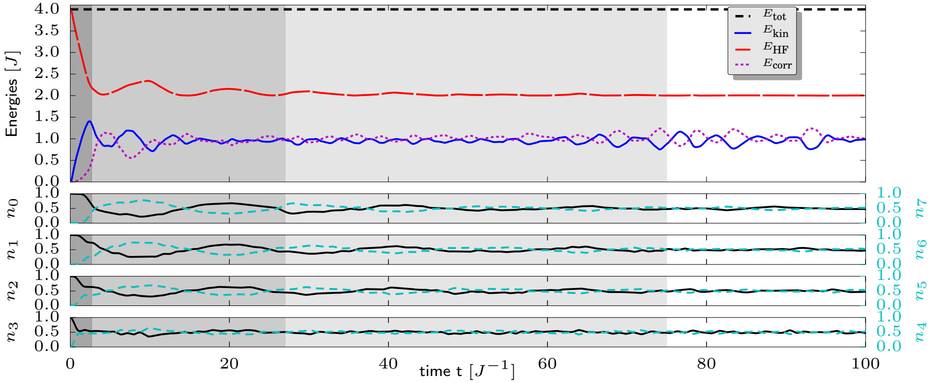
GKBA: correctly describes stages I-II

incorrect: stage III: non-permanent one-particle equilibration

incorrect: stage IV: correlation energy attains negative values

Short-time dynamics ($U = 1.00$): four stages exact calculation: $N = 8$, $n = 1/2$

Sites $0 - 3$ doubly occupied, $4 - 7$ empty



I: $t \leq 3$, ballistic expansion (feature of inhomogeneity)

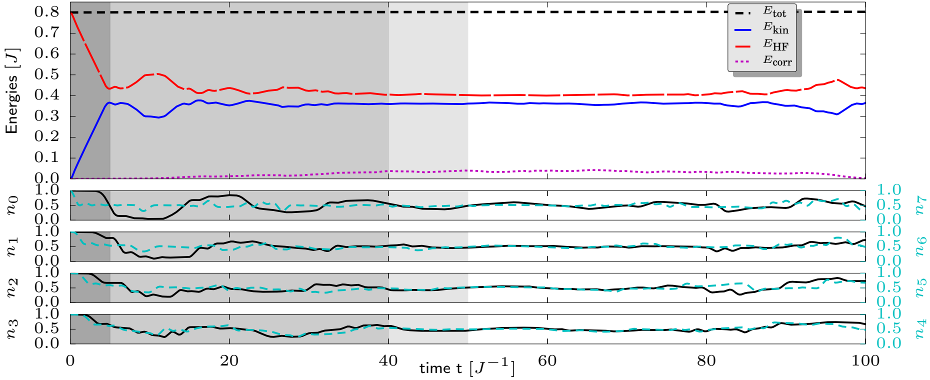
II: $t \leq 25$, correlation build-up/saturation of HF energy

III: $t \leq 75$, one-particle equilibration (occupations)

IV: $t \geq 75$, weak revivals of occupations

GKBA calculation: Noneq. initial state $N = 16,\ n = 1/2,\ U = 0.1$

Sites $0 - 7$ doubly occupied, $8 - 15$ empty



No FCI results possible

Stage II is longer compared to $N = 8$

Relaxation more pronounced for all quantities

Preliminary conclusions from relaxation dynamics

Relaxation dynamics

- 4 Stages of relaxation
- Time-scale of stage I is independent of U but increases with N
- Build-up of correlations faster with larger U in stage **II**
- Time-scale of relaxation of HF-energy mostly independent of U
- Time-scale of stage **III** (relaxation of densities) grows with greater U independent of N
- All quantities show higher degree of relaxation for larger N
- GKBA: for small U very good agreement in stages **I-III**, with larger U only in stages **I-II**

Conclusions and Outlook

Correlated quantum systems in non-equilibrium – Goals:

- self-consistent description of correlation, exchange and nonlinear response to fields, short-time to long-time dynamics

NEGF: can treat mixed and pure states, conserving

1. advantageous scaling with N (limitation: basis size)
2. GKBA $\Rightarrow$ efficiency gain, no artificial damping

Dynamics of finite Hubbard clusters

1. long Hubbard simulations, strong excitation (small U)
2. non-trivial dynamics: four relaxation stages
3. interesting correlation features: stable doublons ($U \gtrsim 3$)
   in progress: T-matrix[4] with GKBA

[4] see also C. Verdozzi, R. van Leeuwen

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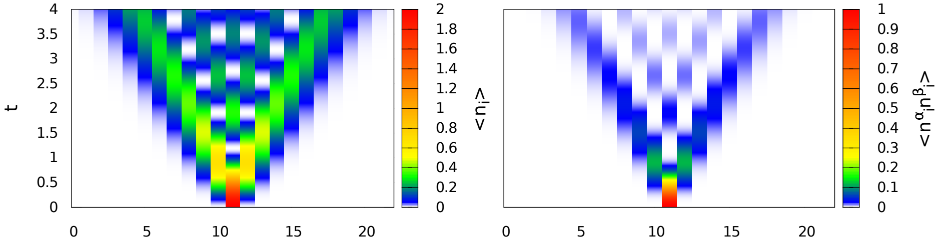
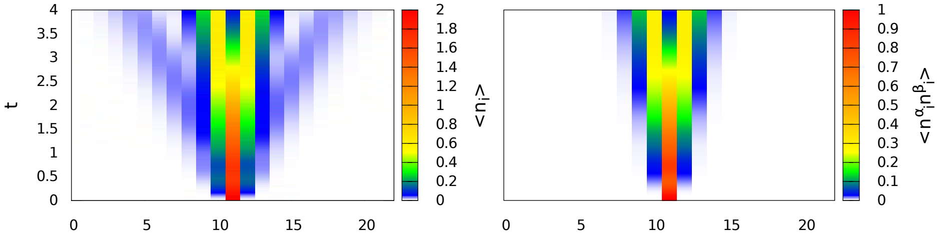
single vs. double occupation, $U = 0$



single vs. double occupation, $U = 7$



Thank you for your attention!

References

- MB and D. Semkat, *Introduction to Computational Methods in Many-Body Physics*, Rinton Press 2006
- K. Balzer, and M. Bonitz, Springer Lecture Notes in Physics **867** (2013)
- www.itap.uni-kiel.de/theo-physik/bonitz/index.html

Postdoc or PhD student position available!

MCTDHF [J. Chem. Phys. **134** 084106 (2011)]

- Time-dependent N-**particle wavefunction** expansion in terms of **Slater determinants**

$$|\Psi(t)\rangle = \sum_{1\leq i_1<\cdots<i_N\leq 2M} C_I(t)\,|\phi_{i_1}(t)\ldots\phi_{i_N}(t)\rangle$$

- $2M < 2N_b$, $N_b \widehat{=}$ one-particle basis dimension, Slater determinant basis reduction by $\binom{2M}{N}$, $M = N/2 \widehat{=}$ Hartree–Fock, $M = N_b \widehat{=}$ Full Configuration Interaction (FCI)
- **Coefficient** and **orbital** equations of motion,

$$\mathrm{i}\frac{\partial}{\partial t}C_I(t) = \sum_J \left\langle I\middle|\,\hat{H}(t)\,\middle|J\right\rangle C_J(t)\,,$$

$$\mathrm{i}\frac{\partial}{\partial t}|\phi_n(t)\rangle = \widehat{\mathbf{P}}\left\{\hat{h}(t)\,|\phi_n(t)\rangle + \sum_{pqrs}\left(\mathbf{D}^{-1}\right)_{np} d_{pqrs}\,\hat{g}_{rs}\,|\phi_q(t)\rangle\right\}\,,$$

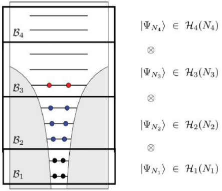
$\mathbf{D} \widehat{=}$ one-particle RDM, $d_{pqrs} \widehat{=}$ two-particle RDM, $\hat{g}_{rs} \widehat{=}$ mean-field integral, projector $\widehat{\mathbf{P}} = \widehat{\mathbf{1}} - \sum_m |\phi_m\rangle\langle\phi_m|$

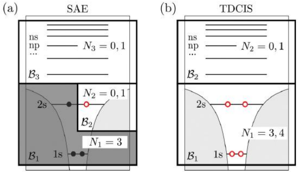
Restricted Active Space TDCI [PRA **86** 053424 (2012)]

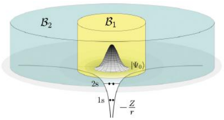
- Time-dependent N-**particle wavefunction** expansion in terms of time-independent **Slater determinants**

$$|\Psi(t)\rangle = \sum_{I\in\Omega} C_I(t)\,|\phi_{i_1}\dots\phi_{i_N}\rangle$$

- Full CI: $\Omega = \{(i_1,\dots,i_\mathsf{N}\,|\,1\le i_1<\dots<i_\mathsf{N}\le N_\mathsf{b})\}$
- Idea of RAS-CI: Drop determinants with minor importance according to physical considerations, e.g., due to energy criterion
- Reduction of full Hilbert space by partitioning and imposing different restrictions in each region





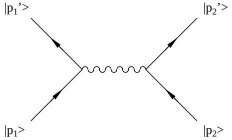


$$\left\{ \frac{\partial}{\partial t} + \frac{\partial E}{\partial \mathbf{p}} \frac{\partial}{\partial \mathbf{R}} - \frac{\partial E}{\partial \mathbf{R}} \frac{\partial}{\partial \mathbf{p}} \right\} f(\mathbf{p}, \mathbf{R}, t) = I(\mathbf{p}, \mathbf{R}, t)$$

$$\begin{aligned} I(\mathbf{p}_1, t) \quad = \quad & \frac{2}{\hbar} \int \frac{d\mathbf{p}_2}{(2\pi\hbar)^3} \frac{d\bar{\mathbf{p}}_1}{(2\pi\hbar)^3} \frac{d\bar{\mathbf{p}}_2}{(2\pi\hbar)^3} \left| \frac{V(\mathbf{p}_1 - \bar{\mathbf{p}}_1)}{\epsilon^{RPA}[\mathbf{p}_1 - \bar{\mathbf{p}}_1, E(\mathbf{p}_1) - E(\bar{\mathbf{p}}_1)]} \right|^2 \\ & \times (2\pi\hbar)^3 \delta(\mathbf{p}_1 + \mathbf{p}_2 - \bar{\mathbf{p}}_1 - \bar{\mathbf{p}}_2) \cdot 2\pi \delta(E_1 + E_2 - \bar{E}_1 - \bar{E}_2) \\ & \times \left\{ \bar{f}_1 \bar{f}_2 (1 \pm f_1)(1 \pm f_2) - f_1 f_2 (1 \pm \bar{f}_1)(1 \pm \bar{f}_2) \right\} |_t \end{aligned}$$

with quasiparticle energy $E_i = E(\mathbf{p}_i)$, $\bar{E}_i = E(\bar{\mathbf{p}}_i)$, $f_i = f(\mathbf{p}_i)$, $\bar{f}_i = f(\bar{\mathbf{p}}_i)$

Example: Quantum Lenard-Balescu (GW) collision integral (Coulomb scattering)



1. Conservation of kinetic (QP) energy, $\frac{d}{dt}\langle E \rangle(t) = 0$
2. Equilibrium solution: Bose/Fermi/Maxwell distribution $\rightarrow$ thermodynamics of ideal gas
3. limited to times larger than correlation time, $t \gg \tau_{corr}$

Generalized quantum kinetic equations [1]

1. Non-Markovian kinetic equations, starting from Bogolyubov hierarchy

   ("top-down", from N-particle density operator)

   Bogolyubov, Klimontovich, Silin, Cassing, ... [2]

2. Second quantization, Nonequilibrium Green functions

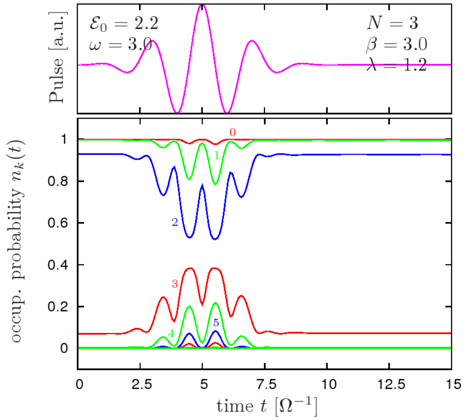
   ("bottom-up", from field operators)

   Bonch-Bruevich, Abrikosov, Keldysh, ...

   Schwinger, Martin, Kadanoff, Baym, Danielewicz, ...

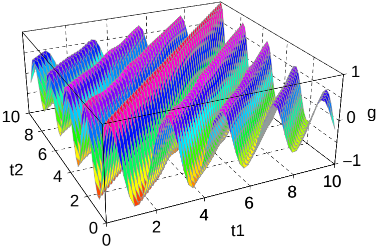
[1] M. Bonitz, *Quantum Kinetic Theory, Teubner 1998*
[2] A. Akbari et al., Phys. Rev. B **85**, 235121 (2012)

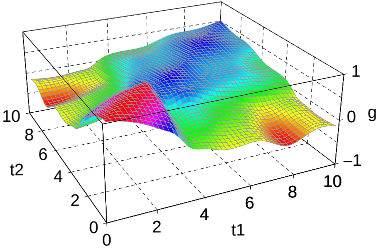
Occupation number dynamics for off-resonant laser excitation, $N = 3$

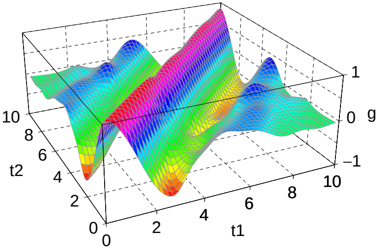


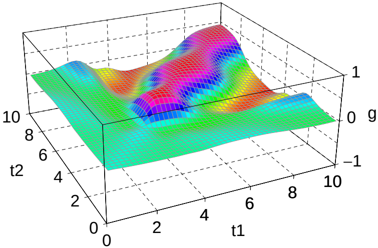
[1] Balzer et al. J. Phys. A **42**, 214020 (2009); Europhys. Lett. **98**, 67002 (2012)

Evolution of Im$g^<$ of levels 1, 2, 3 and 4









Test accuracy of the GKBA using exact propagators.

Example: 2-band semiconductor quantum well, 50fs laser pulse excitation [1]

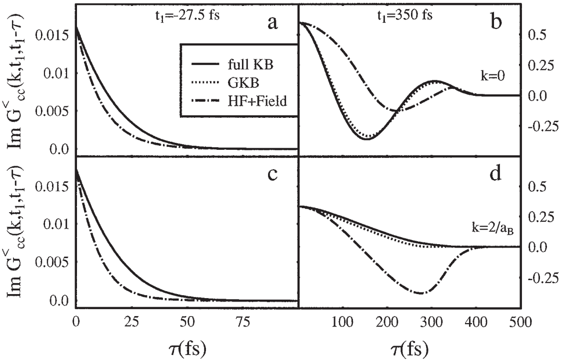


Fig. 2. Imaginary part of $G^<_{cc}(k, t_1, t_2 = t_1 - \tau)$ beginning at the diagonal ($\tau = 0$) and going back in time t_2. The curves correspond to the full two-time result, the GKB ansatz with exact G^r and GKB with G^r in Hartree-Fock approximation. a), c) correspond to $t_1 = -27.5$ fs and b), d) to $t_1 = 350$ fs and to the momenta $k = 0$ (parts a, b) and $k = 2/a_B$ (parts c, d), respectively. The parameters are for case II. At early times, the curves "full KB" and "GKB" are indistinguishable

⇒ GKBA yields accurate and conserving collision rates

[1] N.H. Kwong, M. Bonitz, R. Binder, and H.S. Köhler, phys. stat. sol. (b) **206**, 197 (1998)

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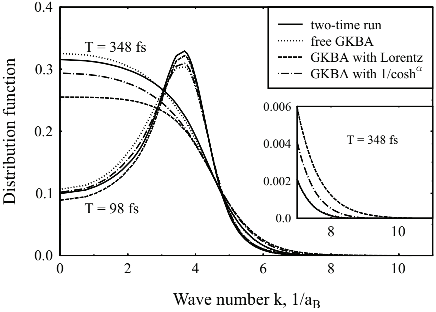
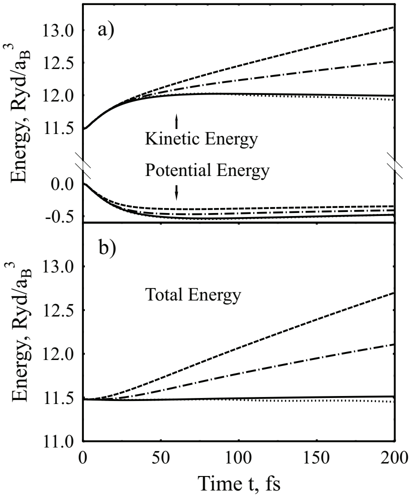
GKBA with damped propagators: thermalization of homogeneous electron gas [1]

1. Lorentzian spectral function:
   $A(p,\tau) = e^{-iE(p)\tau/\hbar} \cdot e^{-\gamma\tau}$
2. Exponential decay vs. frequency:
   $A(p,\tau) = e^{-iE(p)\tau/\hbar} / \cosh^{\alpha} \omega_0 \tau$





⇒ best results for free or HF propagators

[1] M. Bonitz, D. Semkat, and H. Haug, Eur. Phys. J. B **9**, 309 (1999)

1. Second quantization, Nonequilibrium Green functions

   "bottom-up", from field operators $\rightarrow G^{\gtrless}(t,t')$
   GKBA with undamped propagators: $G^{\gtrless}(t,t') = F[\rho(t), \rho(t')]$
   $\rightarrow$ purely single-time theory with NEGF-based approximations (Σ)

2. Non-Markovian quantum kinetic equations

   "top-down", from N-particle density operator $\rho_N(t)$

   $\rho(t) \sim \mathrm{Tr}_{2\dots N}\rho_N(t)$, obeys BBGKY-hierarchy
   approximations: cluster expansion, perturbation theory

NEGF approximations can be identified in BBGKY-eqn. for ρ_2
Selfenergy terms known: follow from eqn. for ρ_3 [1]

[1] M. Bonitz, *Quantum Kinetic Theory*, Teubner 1998
[2] A. Akbari et al., Phys. Rev. B **85**, 235121 (2012)

BBGKY-hierarchy II

$$\begin{aligned} i\hbar\partial_t F_1 - \Big[H_1 , F_1 \Big]_- &= \mathrm{Tr}_2\Big[V_{12} , F_{12} \Big]_- , \\ i\hbar\partial_t F_{12} - \Big[H_{12} , F_{12} \Big]_- &= \mathrm{Tr}_3\Big[V_{13} + V_{23} , F_{123} \Big]_- , \\ \dots \quad \dots \quad \dots \\ F_1(t_0) = F_1^0 , \qquad & F_{12}(t_0) = F_{12}^0 , \qquad \dots \end{aligned}$$

- F_1 coupled to F_{12} etc.
- time-local system of coupled equations
- introduce approximation (decoupling) first, then (anti-)symmetrization
- "intuitive" hierarchy decoupling, $F_{1\dots s} \to 0$, is wrong

BBGKY-hierarchy III – cluster expansion

- Ursell-Mayer expansion:

$$F_{12}(t) = F_1(t)F_2(t) + c_{12}(t)\,,$$
$$F_{123}(t) = F_1(t)F_2(t)F_3(t) + F_1(t)c_{23}(t) + F_2(t)c_{13}(t) + F_3(t)c_{12}(t) + c_{123}(t)\,,$$

- trivial decoupling possible by setting $c_{1\ldots s} \to 0$
- (anti-)symmetrization of operators and hierarchy [1, 2]:

$$F_{12} \longrightarrow F_{12}\Lambda^{\pm}_{12}\,,$$
$$c_{12} \longrightarrow c_{12}\Lambda^{\pm}_{12}\,,$$
$$F_{123} \longrightarrow F_{12}\Lambda^{\pm}_{123}\,,$$
$$c_{123} \longrightarrow c_{123}\Lambda^{\pm}_{123}\,,$$

$$\Lambda^{\pm}_{12}\,|12\rangle = (1 \pm P_{12})\,|12\rangle = |12\rangle \pm |21\rangle\,,$$
$$\Lambda^{\pm}_{123}\,|123\rangle = \Lambda^{\pm}_{12}(1 \pm P_{13} \pm P_{23})\,|123\rangle$$

[1] D.B. Boercker, and J.W. Dufty, Ann. Phys. **119**, 43 (1979)
[2] S. Hermanns, K. Balzer, and M. Bonitz, J. Phys. Conf. Ser. **427**, 012008 (2012)

BBGKY IV – correlation operator hierarchy, $c_{1234=0}$

$$i\hbar\partial_t F_1 - \left[\bar{H}_1^0, F_1\right]_- = \mathrm{Tr}_2\left[V_{12}, c_{12}\right]_- \Lambda_{12}^{\pm}$$

$$i\hbar\partial_t c_{12} - \left[\bar{H}_{12}^0, c_{12}\right]_- = \hat{V}_{12}F_1F_2 - F_1F_2\hat{V}_{12}^{\dagger} + \mathrm{Tr}_3\left[V_{13}+V_{23}, c_{123}\right]_- P_{13;23} + L_{12} + \Pi_{12}$$

$$i\hbar\partial_t c_{123} - \left[\bar{H}_{123}^0, c_{123}\right]_- = \hat{V}_{12}^{\dagger}F_1F_2F_3 + \left(\hat{V}_{13}^{\dagger} + \hat{V}_{23}^{\dagger}\right)F_3c_{12} \mp F_3\left(F_1V_{13} + F_2V_{23}\right)c_{12} \mp \left(c_{13}V_{13} + c_{23}V_{23}\right)c_{12} + (1\to 2\to 3) + \Pi_{123} + L_{123} - \text{h.c.(rhs.)}$$

$$\bar{H}_1^0 = H_1 + U_1^{\mathrm{HF}}, \qquad U_1^{\mathrm{HF}} = \mathrm{Tr}_2 V_{12}F_2\Lambda_{12}^{\pm}$$

$$\bar{H}_{1\ldots s}^0 = \bar{H}_1^0 + \ldots \bar{H}_s^0, \qquad \hat{V}_{12} = (1 \pm F_1 \pm F_2)V_{12}$$

ladder terms : $L_{12} = \hat{V}_{12}c_{12} - c_{12}\hat{V}_{12}^{\dagger}$, (T − matrix)

polarization terms : $\Pi_{12} = \mathrm{Tr}_3\left[V_{13}\Lambda_{13}^{\pm}, F_1\right]_- c_{23}\Lambda_{23}^{\pm}$, (Balescu, GW)

selfenergy terms : renormalize $\bar{H}_{12}^0$, exactly recover Full GKBA

$$F_1(t_0) = F_1^0,\ c_{12}(t_0) = c_{12}^0,\ c_{123}(t_0) = c_{123}^0,\ P_{13;23} = (1 \pm P_{13} \pm P_{23})$$

M. Bonitz, *Quantum Kinetic Theory*; S. Hermanns, K. Balzer, and M. Bonitz, J. Phys. Conf. Ser. **427**, 012008 (2012)

Born approx.: Non-Markovian Landau equation

Spatially homogeneous system. Direct 2nd Born approximation

$$\frac{\mathrm{d}}{\mathrm{d}t}F_1(t) = \frac{2\mathcal{V}^2}{\bar{h}^2}\int_0^{t-t_0}\mathrm{d}\tau\int\frac{\mathrm{d}\boldsymbol{p}_2}{(2\pi\bar{h})^3}\int\frac{\mathrm{d}\bar{\boldsymbol{p}}_1}{(2\pi\bar{h})^3}\int\frac{\mathrm{d}\bar{\boldsymbol{p}}_2}{(2\pi\bar{h})^3}\,(2\pi\bar{h})^3\delta(\boldsymbol{p}_{12}-\bar{\boldsymbol{p}}_{12})$$
$$\times\, V^2\left(\frac{\bar{\boldsymbol{p}}_1-\boldsymbol{p}_1}{\bar{h}}\right)\cos\left\{\frac{E_{12}-\bar{E}_{12}}{\bar{h}}\tau\right\}\mathrm{e}^{-(\gamma_{12}+\bar{\gamma}_{12})\tau/\bar{h}}$$
$$\times\left\{\bar{F}_1\bar{F}_2[1-F_1][1-F_2]-F_1F_2[1-\bar{F}_1][1-\bar{F}_2]\right\}\Big|_{t-\tau}$$

- $\mathbf{p}_{12}=\mathbf{p}_1+\mathbf{p}_2, \quad E_{12}=E_1+E_2, \quad \gamma_{12}=\gamma_1+\gamma_2, \quad \gamma_1=\mathrm{Im}\Sigma^R(p_1)$
- Special cases:

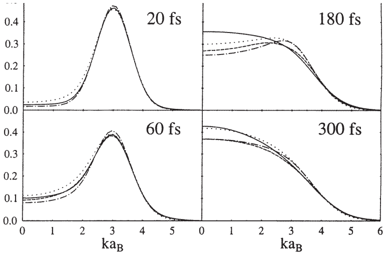
a) free GKBA: $\gamma_i\to 0$;
b) neglect of retardation: $F(t-\tau)\to F(t) \quad \Rightarrow \text{integrand}\sim\mathrm{sinc}(E_{12}-\bar{E}_{12})$
c) Markov limit: $t_0\to-\infty, \Rightarrow \text{integrand}\sim\delta(E_{12}-\bar{E}_{12})$

M. Bonitz, S. Köhler et al. J. Phys. Cond. Matt. **8**, 6057 (1996)

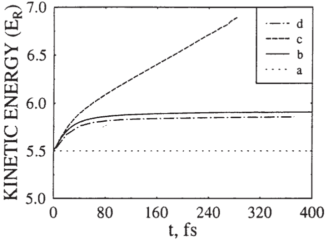
Relaxation of Laser-excited homogeneous e-h-plasma

Coulomb scattering in Born approximation (Yukawa potential):

Electron momentum distribution



Mean kinetic energy

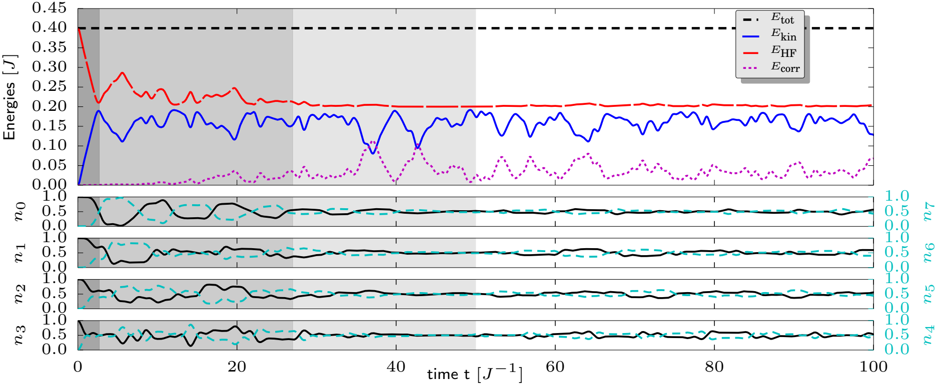


- from fastest to slowest: no retard. (b), Markov limit (a), free GKBA (c), KBE (d)
- b, c, d conserve total energy [kinetic energy in (c) incorrect]

M. Bonitz, S. Köhler et al. J. Phys. Cond. Matt. **8**, 6057 (1996)

Short-time dynamics ($U = 0.75$): four stages exact calculation: $N = 8$, $n = 1/2$

Sites $0 - 3$ doubly occupied, $4 - 7$ empty



I: $t \leq 3$, ballistic expansion (feature of inhomogeneity)

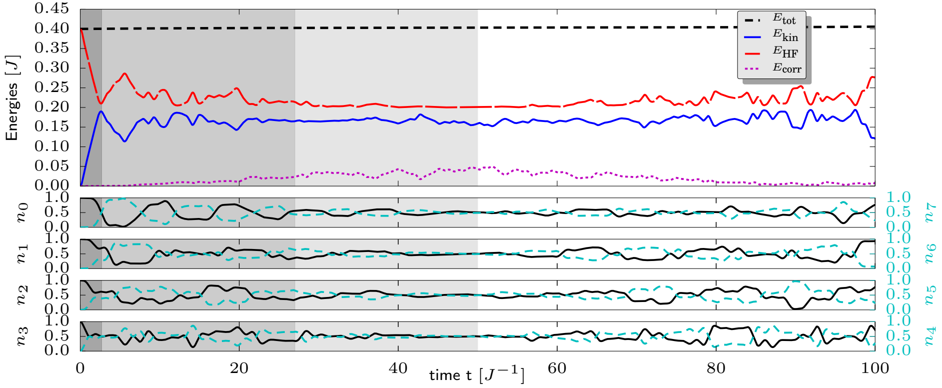
II: $t \leq 25$, correlation build-up

III: $t \leq 50$, one-particle equilibration (occupations)

IV: $t \geq 50$, weak revivals of occupations

GKBA calculation: Noneq. initial state $N = 8,\ n = 1/2,\ U = 0.75$

Sites $0 - 3$ doubly occupied, $4 - 7$ empty



GKBA: correctly describes time-scales of stages I-III
shows incorrect return to non-equilibrated state

For large U: pertubation theory in $1/U \rightarrow$ effective Hamiltonian describing quasiparticles called 'doublons':

$$\hat{\mathcal{H}}_{eff} = \frac{J^2}{U} \sum_{\langle i,j \rangle} \hat{d}_i^\dagger \hat{d}_j + \sum_{ij} V_{ij} \hat{n}_i^d \hat{n}_j^d$$

$V_{ij} = \infty$ for $i = j$, $V_{ij} = -\frac{J^2}{U}$ for $ij = \langle i, j \rangle$, $\hat{d}_i^\dagger := \hat{c}_{i\uparrow}^\dagger \hat{c}_{i\downarrow}^\dagger$, $\hat{n}_i^d := \hat{d}_i^\dagger \hat{d}_i$

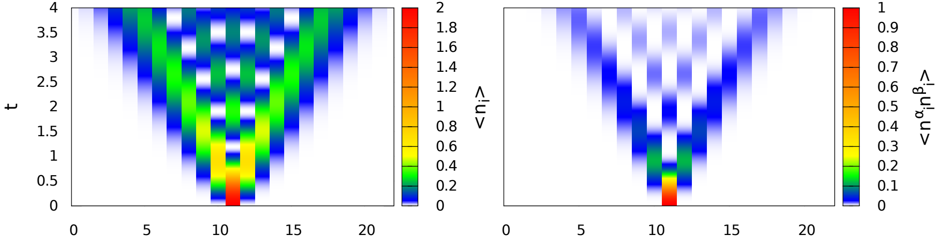
Previous studies of expansion dynamics and stability of doublons:

- Ronzheimer *et al.*, Phys. Rev. Lett. **110**, 205301 (2013)
- Hoffmann *et al.*, Phys. Rev. B **86**, 205127 (2012)

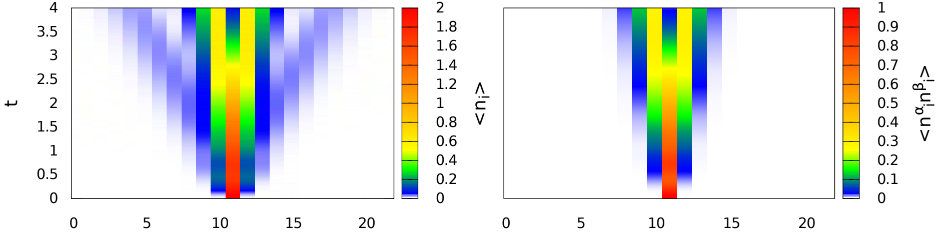
Goal here: study simple exactly solvable (Full TDCI) model: $N = 2$ fermions initially localized at $i_0 = 11$, $N_\mathrm{b} = 23$.

$t = 0$: 1 doublon in center. Expansion dynamics

single vs. double occupation, $U = 0$



single vs. double occupation, $U = 7$

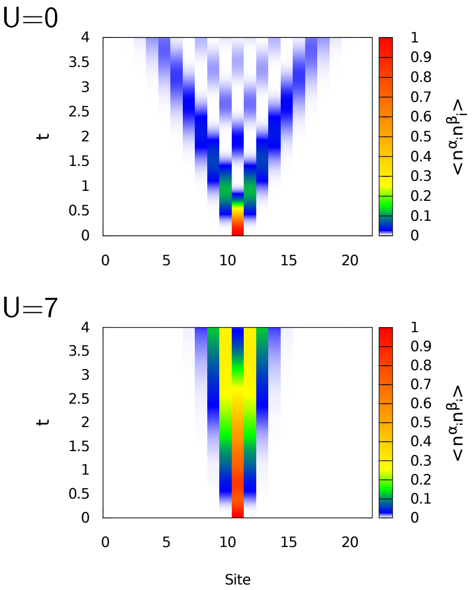


Quantifying the expansion dynamics

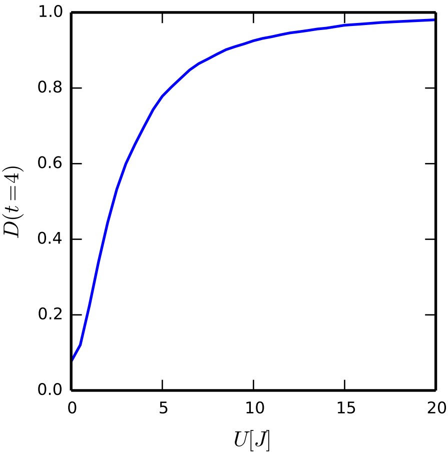
- total double occupancy: $D(t) := \sum_i \left\langle \hat{n}_i^{\uparrow} \hat{n}_i^{\downarrow} \right\rangle$
- width of probability cloud: $R^2(t) := \frac{1}{N} \sum_i \langle \hat{n}_i \rangle (i - i_0)^2$
- expansion velocity[5]: $v_{\mathrm{r}} := \frac{d}{dt} \sqrt{R^2(t) - R^2(0)}$
- width of doublon probability cloud: $R_D^2(t) := \frac{1}{D(t)} \sum_i \left\langle \hat{n}_i^{\uparrow} \hat{n}_i^{\downarrow} \right\rangle (i - i_0)^2$
- doublon expansion velocity: $v_{\mathrm{r,Doublon}} := \frac{d}{dt} \sqrt{R_D^2(t) - R_D^2(0)}$

[5] compare Ronzheimer *et al.*, Phys. Rev. Lett. **110**, 205301 (2013)

Total double occupancy $D(t;U)$

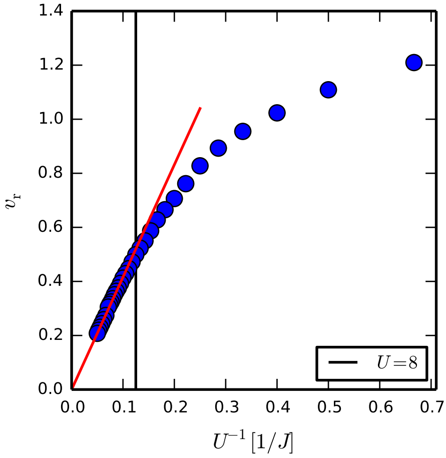


$$D(t) := \sum_i \left\langle \hat{n}_i^{\uparrow} \hat{n}_i^{\downarrow} \right\rangle$$



Total vs. doublon expansion velocity

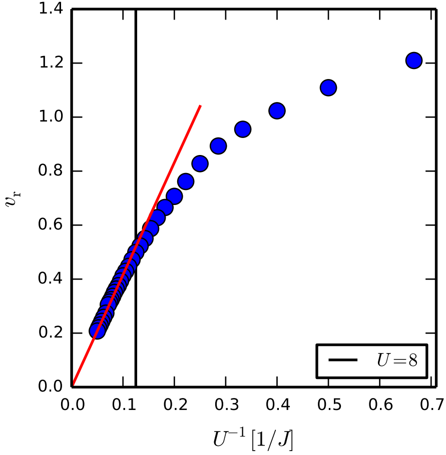
$$v_{\mathrm{r}} := \frac{d}{dt}\sqrt{R^2(t) - R^2(0)}$$

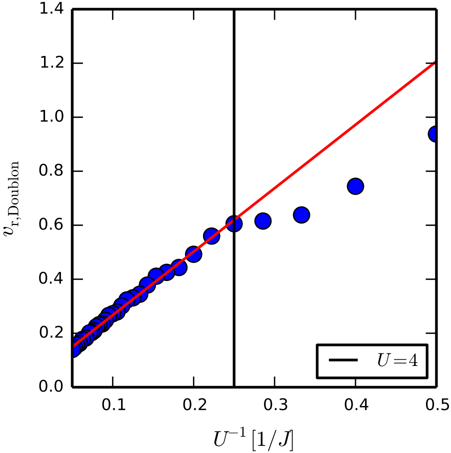


Total vs. doublon expansion velocity

$$v_{\mathrm{r}} := \frac{d}{dt}\sqrt{R^2(t) - R^2(0)}$$

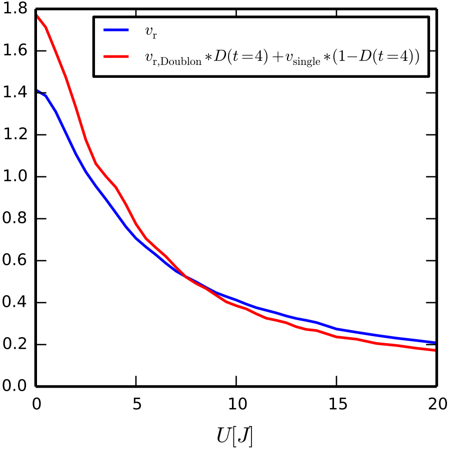
$$v_{\mathrm{r,Doublon}} := \frac{d}{dt}\sqrt{R_D^2(t) - R_D^2(0)}$$





Two-fluid model

Define $v_{\text{single}} := \left[i_0 - \frac{\sum_{i=0}^{7} \langle \hat{n}_i \rangle i}{\sum_{i=0}^{7} \langle \hat{n}_i \rangle} \right] t^{-1}, \quad t = 4$



- $U \gtrsim 8$: doublon stable, dynamics described by effective Hamiltonian
- $3 \lesssim U \lesssim 8$: two-fluid model for expansion dynamics

  in agreement with Kajala *et al.*, Phys. Rev. Lett. **106**, 06401 (2011)
- $U \lesssim 3$: effective Hamiltonian not applicable