Femtosecond correlated fermion dynamics in finite quantum systems

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in collaboration with:
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Motivation

Our main interest: correlated charged particle systems in and out of equilibrium

Methods:

- Kinetic theory for plasmas (PIC-MCC), MD for surface processes
- *ab initio* thermodynamics for warm dense matter (quantum Monte Carlo, avoid sign problem, f_{xc}^{UEG} with 0.3% accuracy)[1], functional available in library *libxc*
- nonequilibrium Green functions (NEGF) approach to inhomogeneous systems

Recent nonequilibrium applications with NEGF:

- Time-resolved photoionization of few-electron atoms and molecules
- Dynamics of finite Hubbard clusters following a confinement quench
- Interaction of low-temperature plasmas with solids: ion stopping, electronic correlation effects, doublon formation
- Photoexcitation dynamics of graphene nanoribbons. Carrier multiplication

[1] Schoof *et al.* PRL 2015, Dornheim *et al.*, PRL 2016, Groth *et al.*, PRL 2017, Physics Reports 2018

Finite correlated quantum systems

Fermionic atoms in optical lattices
tunable lattice depth and interaction

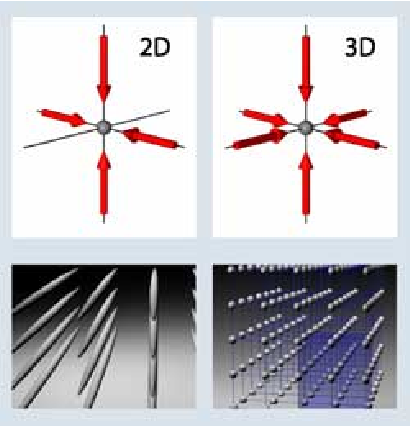
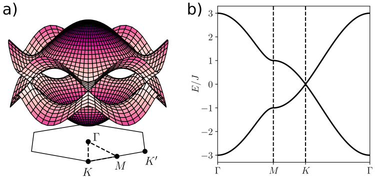
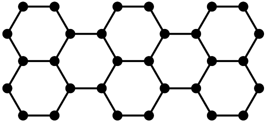


Fig.: M. Greiner (Harvard)

Graphene: high mobility, no bandgap



Graphene nanoribbons:
finite tunable bandgap



Outline

Christian-Albrechts-Universität zu Kiel
Mathematisch-Naturwissenschaftliche Fakultät

1. Nonequilibrium Green functions (NEGF)
   - Theoretical framework
   - Strong electronic correlations. Hubbard model
2. NEGF results for 2D cold atom expansion

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

Keldysh Green functions (NEGF)

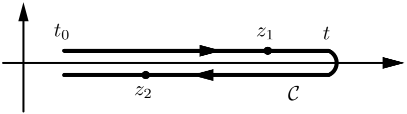
two times $z, z' \in \mathcal{C}$ ("Keldysh contour"), arbitrary one-particle basis $|\phi_i\rangle$

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

average with ρ^N

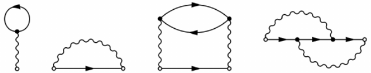
Keldysh–Kadanoff–Baym equations (KBE) on $\mathcal{C}$ (2×2 matrix):

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



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

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



Real-time Keldysh-Kadanoff-Baym equations

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

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

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

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

- Propagators (spectral properties)

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

- Correlation functions $G^{\gtrless}$ (statistical properties) obey real-time KBE

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

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

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])

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

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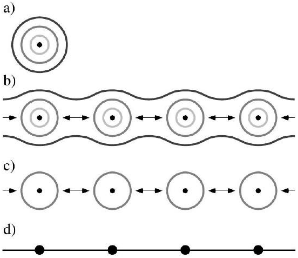
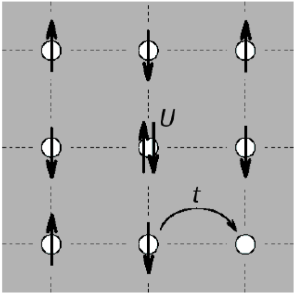


$\Rightarrow$



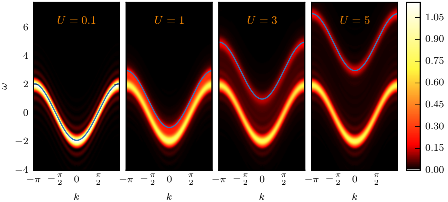
$$\hat{H}(t) = J \sum_{ij,\,\alpha} h_{ij}\, \hat{c}^{\dagger}_{i\alpha}\hat{c}_{j\alpha} + U \sum_i \hat{c}^{\dagger}_{i\uparrow}\hat{c}_{i\uparrow}\hat{c}^{\dagger}_{i\downarrow}\hat{c}_{i\downarrow} + \sum_{ij,\alpha\beta} f_{ij,\alpha\beta}(t)\, \hat{c}^{\dagger}_{i\alpha}\hat{c}_{j\beta}$$

$h_{ij} = -\delta_{\langle i,\, j\rangle}$ and $\delta_{\langle i,\, j\rangle} = 1$, if (i, j) nearest neighbors, $\delta_{\langle i,\, j\rangle} = 0$ otherwise;
on-site repulsion $(U > 0)$ or attraction $(U < 0)$, U favors *doublons* (correlations)

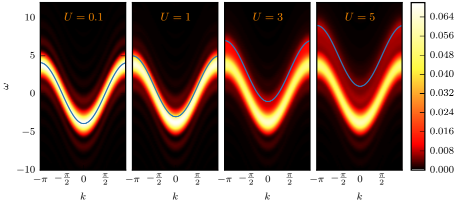
- f: excitation (1-particle hamiltonian): EM field, quench, particle impact etc.
- finite inhomogeneous system, size and geometry dependence

Dispersion relation

1D:



2D:



- spectral function (peaks yield single-particle dispersion)

$$A(\omega, \boldsymbol{k}) \quad = \quad \frac{\mathrm{i}\hbar}{N_s N_t} \sum_{\boldsymbol{s}\boldsymbol{s'}tt'} \mathrm{e}^{-\mathrm{i}\boldsymbol{k}(\boldsymbol{s}-\boldsymbol{s'})} \mathrm{e}^{-\mathrm{i}\omega(t-t')} \left[G^{>}_{\boldsymbol{s}\boldsymbol{s'}}(t,t') - G^{<}_{\boldsymbol{s}\boldsymbol{s'}}(t,t')\right]$$

- upper band: doublons, upshifted by $\sim U$
- KBE result for local doublon occupation (two-particle quantity):

$$d_i(t) = \langle \hat{n}_{i\uparrow}(t)\hat{n}_{i\downarrow}(t)\rangle = -\frac{\mathrm{i}\hbar}{U} \sum_k \int_{\mathcal{C}} ds\, \Sigma_{ik}(t,s) G_{ki}(s,t)$$

$$d_i^{\mathrm{HF}}(t) = n_{i\uparrow}(t)\, n_{i\downarrow}(t)$$

- correlations: clearly visible in deviations from Hartree-Fock

Selfenergy approximations[2]

Choice depends on coupling strength, density (filling)

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

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

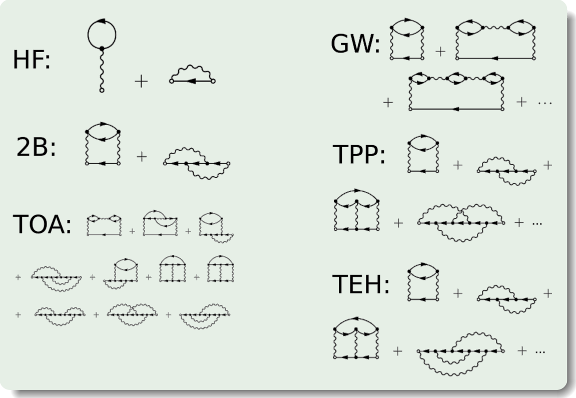
GW: ∞ bubble summation, dynamical screening effects

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

electron-hole T-matrix (TEH): ∞ ladder sum in ph channel

FLEX (GW+TPP+TEH)

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



[2]Conserving, nonequilibrium $\Sigma(t, t')$, applies for ultra-short to long times

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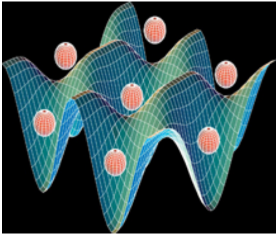
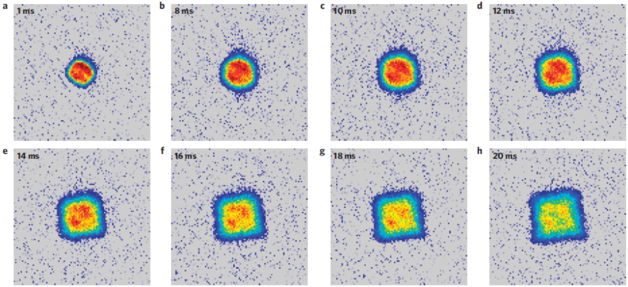
Time-resolved expansion of fermionic atoms

nature physics ARTICLES
PUBLISHED ONLINE: 15 JANUARY 2012 | DOI: 10.1038/NPHYS2205

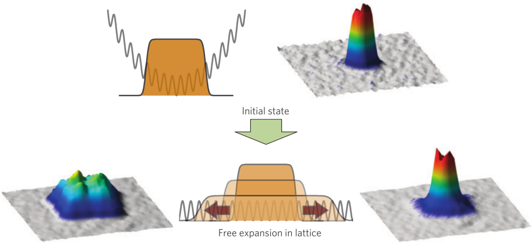
Fermionic transport and out-of-equilibrium dynamics in a homogeneous Hubbard model with ultracold atoms

Ulrich Schneider[1,2]*, Lucia Hackermüller[1,3], Jens Philipp Ronzheimer[1,2], Sebastian Will[1,2], Simon Braun[1,2], Thorsten Best[1], Immanuel Bloch[1,2,4], Eugene Demler[5], Stephan Mandt[6], David Rasch[6] and Achim Rosch[6]





- 2D optical lattice, ca. 200 000 atoms
- atom-atom interaction strength tuned (via Feshbach resonance)
- t<0: confinement in trap center, doubly occupied lattice sites
- t=0: confinement rapidly removed ("quench"):
  system far from equilibrium ⇒ start of diffusion, equilibration



- at strong coupling: center ("core") does not expand due to **doublon** formation

A challenge for quantum many-body dynamics...

Quote from Schneider *et al.*, (p. 216):

"Although the expansion can be modelled in 1D (...) using DMRG methods (...), so far no methods are available to calculate the dynamics quantum-mechanically in higher dimensions"

Similar claims in many experimental papers, for example:

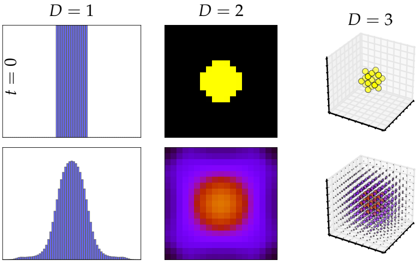
"Quantengase unter dem Mikroskop", M. Greiner, I. Bloch, Phys. Journal Okt. 2015:

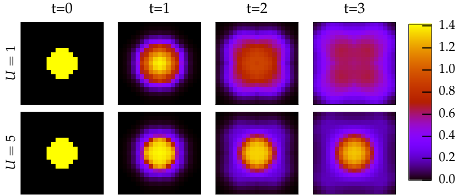
"*Ein anderes Gebiet, in dem Experimente schon heute leistungsfähiger als Computersimulationen sind, ist die Untersuchung von Nichtgleichgewichtsprozessen in Quanten-Vielteilchensystemen ... bisherige Algorithmen auf eindimensionale Systeme beschränkt sind und meistens nur die Dynamik für sehr kurze Zeiten berechnen können.*"

Not exactly true...

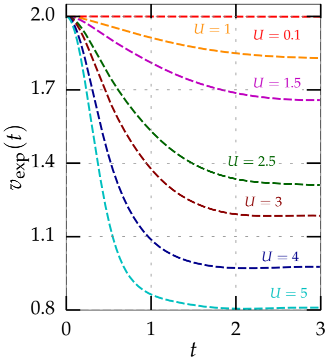
NEGF results for fermion expansion

- $t = 0$: central array of doubly occupied sites
- confinement quench initiates expansion
- expansion speed, dynamics time-dependent, depend on
  - dimensionality D, interaction strength U, particle number N





Time evolution of the expansion velocity[3]



Diffusion quantities

- **mean squared displacement**

$$R^2(t) = \frac{1}{N} \sum_{\boldsymbol{s}} n_{\boldsymbol{s}}(t)[\boldsymbol{s} - \boldsymbol{s}_0]^2$$

  $\boldsymbol{s}_0$: center of the system

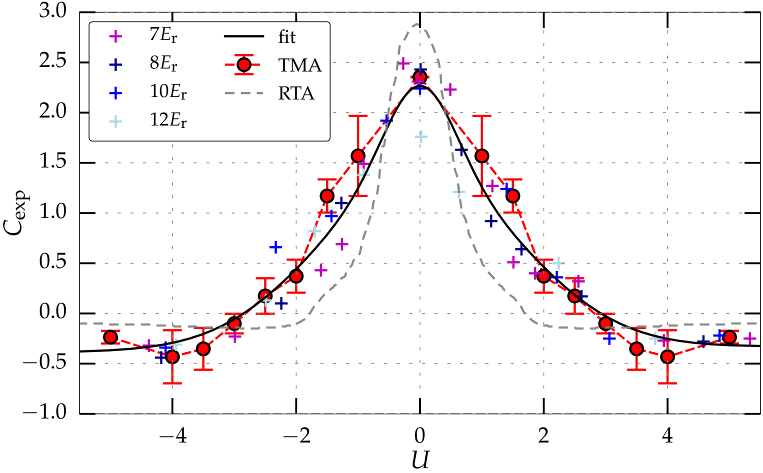
- rescaled cloud **diameter** $d(t) = \sqrt{R^2(t) - R^2(0)}$
- **expansion velocity** $v_{\mathrm{exp}}(t) = \frac{\mathrm{d}}{\mathrm{d}t} d(t)$
- **asymptotic expansion velocity**

$$v_{\mathrm{exp}}^{\infty} = \lim_{t\to\infty} v_{\mathrm{exp}}(t)$$

- example: $N = 58$ doubly occupied sites in 2D
- perform extrapolation with respect to N
- similar procedure for "core expansion velocity" ($\sim$ FWHM)

[3] N. Schlünzen, S. Hermanns, M. Bonitz, and C. Verdozzi, Phys. Rev. B **93**, 035107 (2016)

NEGF result[4] vs. experiment and RTA[5]



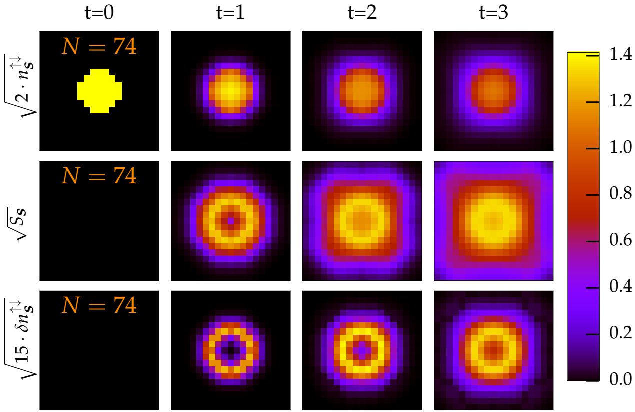
- agreement with measurements for the *final stage* of the dynamics
- in addition: NEGF predict early stages, correlation dynamics etc.

[4] 2-time T-matrix, N. Schlünzen, S. Hermanns, M. Bonitz, and C. Verdozzi, Phys. Rev. B **93**, 035107 (2016)

[5] U. Schneider *et al.*, Nature Physics **8**, 213-218 (2012)

Site-resolved evolution of correlations

- double occupation $n_{\boldsymbol{s}}^{\uparrow\downarrow}$
- local entanglement entropy $S_{\boldsymbol{s}}$
- pair correlation function $\delta n_{\boldsymbol{s}}^{\uparrow\downarrow} = n_{\boldsymbol{s}}^{\uparrow\downarrow} - n_{\boldsymbol{s}}^{\uparrow} n_{\boldsymbol{s}}^{\downarrow}$



- insights into the early expansion phase
- measurable in quantum atom microscopes

Conclusions[6]

- first correlated quantum dynamics simulations in 2D. Excellent agreement with experiments
- **NEGF** well suited to describe **nonequilibrium dynamics in correlated finite (inhomogeneous) systems**, quantitatively reliable, predictive power
- controlled choice of selfenergy: dictated by filling and interaction strength, presently accurate up to $U \simeq$ bandwidth
- NEGF not restricted by geometry, dimensionality or ensemble. Approximately cubic scaling with N_B, T
- Current applications: **2D graphene clusters, nanoribbons**: promising electronic and optical properties due to correlations: ion stopping, carrier multiplication, photon sidebands

[6] M. Bonitz, *Quantum Kinetic Theory*, 2nd ed. Springer 2016
K. Balzer, M. Bonitz, *Nonequilibrium Green Functions Approach to Inhomogeneous systems*, Springer 2013
www.itap.uni-kiel.de/theo-physik/bonitz