Nonequilibrium Green's functions approach to the sub-femtosecond dynamics of plasmas and atoms in intense x-ray fields

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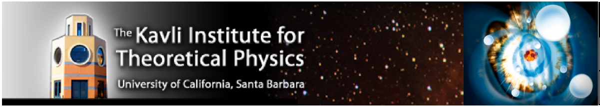


C A U
Christian-Albrechts-Universität zu Kiel
Mathematisch-Naturwissenschaftliche Fakultät

KITP, X-Ray Frontiers

Wednesday, August 25 2010—2:00 pm

Where is Kiel?

Figure: Kiel

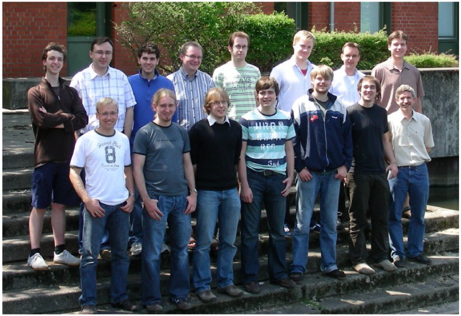
Where is Kiel?

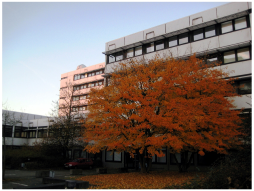
Figure: Kiel

What is Kiel?

Figure: Picture of the "Kieler Woche" 2008

Chair Statistical Physics





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Back from left: Gabriel Dominique-Marleau*, Alexei Filinov, Paul LaPlante*, Henning Baumgartner, Hanno Kählert, Lasse Rosenthal, Patrick Ludwig, Jens Böning

Front: Christian Henning, Karsten Balzer, Sebastian Bauch, Torben Ott, Martin Heimsoth, David Hochstuhl, MB

missing: Hauke Thomsen, Henning Bruhn

* Guest students participating DAAD-RISE-summer internship 2009

Main Research Directions

Classical plasmas
- plasma crystals
- fluid theory
- dusty plasmas

Atoms in strong fields
- correlation dynamics
- photoionization
- (sub-)fs dynamics

Dense quantum plasmas
- planets, dwarf stars
- laser plasmas
- warm dense matter

Strongly correlated Coulomb systems
- many-body effects
- first-principle simulations

Electrons in quantum dots
- Wigner crystallization

Quark-Gluon plasma
- equation of state
- transport properties

Nonideal bosons
- excitons in bilayers
- atoms in traps
- Bose condensation, superfluidity

Outline

Outline

Why Nonequilibrium Green's Functions (NEGFs)?

- Interacting many-body system in strong ultrashort fields
- Self-consistent treatment of e-e correlations and nonlinear field effects without perturbation approximation
- Conservation laws guaranteed
- Dynamics on (sub)fs time scales
- Systematic approach to approximations (Feynman diagrams)

Applications

- Condensed matter systems
- Plasmas
- Electrons in atoms, molecules
- Nuclear matter, high energy physics
- Coupled dynamics of electrons, plasmons and photons
- . . .

NEGFs are the most systematic starting point to these problems:

(i) Consistent derivation of approximate theories

(ii) Direct numerical analysis

Outline

1 Introduction

2 Many-electron problem. Nonequilibrium Green's functions
- Second quantization. NEGFs
- Keldysh-Kadanoff-Baym equations (KKBE)

3 Applications
- Homogeneous Coulomb systems. Plasmas
- Model atoms and molecules

4 Summary

Hamiltonian in second quantization

- **Hamiltonian** for interacting system

$$H(t) = \int \mathrm{d}^3 r\, \Psi_H^\dagger(\boldsymbol{r},t) H_1(\boldsymbol{r},t) \Psi_H(\boldsymbol{r},t) + \frac{1}{2} \iint \mathrm{d}^3 r\, \mathrm{d}^3 r'\, \Psi_H^\dagger(\boldsymbol{r},t)\Psi_H^\dagger(\boldsymbol{r}',t) H_2(\boldsymbol{r}-\boldsymbol{r}') \Psi_H(\boldsymbol{r}',t)\Psi_H(\boldsymbol{r},t)$$

e.g. $H_1(\boldsymbol{r},t) = [\boldsymbol{p} - eA(\boldsymbol{r},t)]^2 + \phi(\boldsymbol{r},t),$ $\quad H_2(\boldsymbol{r}-\boldsymbol{r}') = e^2|\boldsymbol{r}-\boldsymbol{r}'|^{-1}$

- **Field operators** (Heisenberg picture) Ψ, $\Psi^\dagger$ with commutation relation for bosons $(-)$, anti-commutation relation for fermions $(+)$

$$\left[\Psi^{(\dagger)}(\boldsymbol{r},t), \Psi^{(\dagger)}(\boldsymbol{r}',t)\right]_\mp = 0 \qquad \left[\Psi(\boldsymbol{r},t), \Psi^\dagger(\boldsymbol{r}',t)\right]_\mp = \delta(\boldsymbol{r}-\boldsymbol{r}')$$

$\Rightarrow$ Theory has "built in" spin statistics
Symmetry/anti-symmetry of N-particle states exactly garanteed

Macroscopic observables

- **Equilibrium ensemble average**. Density operator

$$\langle O\rangle = \mathrm{Tr}\{\rho O\} \qquad \rho = \frac{1}{Z}\exp\{-(\beta H - \mu N)\}$$

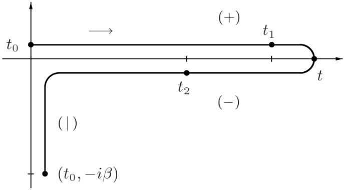
- **Nonequilibrium expectation values**. Switch on the perturbing field at $t = t_0$, $\langle O\rangle \to \langle O\rangle(t)$, use Heisenberg operators

$$\langle O\rangle(t) = \frac{\mathrm{Tr}\{e^{\beta\mu N} U(t_0 - i\beta, t_0) U(t_0, t) O U(t, t_0)\}}{\mathrm{Tr}\{e^{\beta\mu N} U(t_0 - i\beta, t_0)\}}$$

with evolution operator ($\hbar = 1$)

$$U(t, t_0) = T\exp\left(-i\int_{t_0}^{t} d\bar{t}\, H(\bar{t})\right) \qquad U(t_0 - i\beta, t_0) = e^{-\beta H(t_0)}$$

- Time-evolution runs along Keldysh contour[1]
$\mathcal{C} = \{t \in \mathbb{C} | \mathfrak{Re}\, t \in [t_0, \infty], \mathfrak{Im}\, t \in [0, -\beta]\}$



[1] L.V. Keldysh, Sov. Phys. JETP **20**, 1018 (1965)

Definition of Nonequilibrium Green's functions

- **Replace** $\langle O \rangle \to \langle \Psi_H \Psi_H^\dagger \rangle$: 1-particle Green's function

$$G(\boldsymbol{r},t;\boldsymbol{r}',t') = \pm \mathrm{i} \left\langle T_{\mathcal{C}} \Psi_H(\boldsymbol{r},t) \Psi_H^\dagger(\boldsymbol{r}',t') \right\rangle$$

  $T_{\mathcal{C}}$: time-ordering along $\mathcal{C}$
  t, t' belong to one of 3 branches $\Rightarrow$ G is 3×3 matrix

- **Keldysh Green's functions** (matrix elements on $\mathcal{C}$). Denote $1 = (t, \boldsymbol{r}, s_z)$

  1. Correlation functions: $G^<(1,1') = \mp \mathrm{i} \langle \Psi_H^\dagger(1') \Psi_H(1) \rangle$
     $G^>(1,1') = -\mathrm{i} \langle \Psi_H(1) \Psi_H^\dagger(1') \rangle$
  2. Retarded/Advanced functions:
     $$G^{\mathrm{R/A}}(1,1') = \pm \Theta[\pm(t-t')] \left\{ G^>(1,1') - G^<(1,1') \right\}$$
  3. Real branch - imaginary branche components: $G^{\rceil}(1,1')$, $G^{\lceil}(1,1')$
  4. Imaginary branche component: Matsubara (equilibrium) Green's function: $G^M(1,1')$

Information contained in the NEGF

- **Physical content**: Time-dependent macroscopic observables

$$\langle O\rangle(t) = \int \mathrm{d}^3r\, \left\{O(\boldsymbol{r}',t)\left\langle \Psi_H^\dagger(\boldsymbol{r},t)\Psi_H(\boldsymbol{r}',t)\right\rangle\right\}_{\boldsymbol{r}=\boldsymbol{r}'}$$
$$= \pm\mathrm{i}\int \mathrm{d}^3r\, \left\{O(\boldsymbol{r}',t)G^<(\boldsymbol{r},t;\boldsymbol{r}',t)\right\}_{\boldsymbol{r}=\boldsymbol{r}'}$$

  - Particle density: $n(\boldsymbol{r},t) = \pm\mathrm{i}G^<(\boldsymbol{r},t;\boldsymbol{r},t)$
  - Density matrix: $F(\boldsymbol{r},\boldsymbol{r}';t) = \pm\mathrm{i}G^<(\boldsymbol{r},t;\boldsymbol{r}',t')|_{t'=t}$
  - Current density: $\boldsymbol{j}(\boldsymbol{r},t) = \pm\mathrm{i}\left\{\frac{\nabla-\nabla'}{2\mathrm{i}m}G^<(\boldsymbol{r},t,\boldsymbol{r}',t)\right\}_{\boldsymbol{r}'=\boldsymbol{r}}$
  - Interaction energy also follows from the 1-particle Green's function[1]:

$$\langle H_{\mathrm{int}}\rangle(t) = \pm\frac{V}{4}\int\frac{\mathrm{d}^3p}{(2\pi\hbar)^3}\left\{[\mathrm{i}\partial_t - \mathrm{i}\partial_{t'}] - \frac{\boldsymbol{p}^2}{m}\right\}G^<(\boldsymbol{p},t,t')|_{t=t'}$$

[1] L.P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, New York, 1962)

Information contained in the NEGF (cont.)

- Wigner function

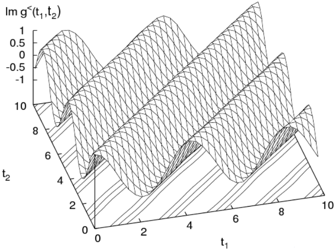
$$f(\boldsymbol{p}, \boldsymbol{R}, T) = \pm \frac{\mathrm{i}}{2\pi} \int \mathrm{d}\omega \, G^{<}(\boldsymbol{p}, \omega, \boldsymbol{R}, T)$$

  with relative (CoM) quantities $\bar{x} = x - x'$ ($X = \frac{1}{2}(x + x')$), and Fourier transform $\bar{\boldsymbol{r}} \to \boldsymbol{p}$ and $\bar{t} \to \omega$

- Single particle spectrum (spectral function)

$$a(\omega, \boldsymbol{R}, \boldsymbol{p}, T) = \mathrm{i} \int \mathrm{d}\bar{t} \, \mathrm{e}^{\mathrm{i}\omega\bar{t}} \left\{ G^{>} - G^{<} \right\} (\boldsymbol{R}, \boldsymbol{p}, T + \bar{t}/2, T - \bar{t}/2)$$

- Nonequilibrium density of states $\rho(\omega) = \mathrm{Tr} \left\{ a(\omega, \boldsymbol{R}, \boldsymbol{p}, T) \right\}$



Equations of motion for Keldysh Green's functions

- Use Heisenberg's equation of motion: $\mathrm{i}\,\partial_t\,\Psi_H^{(\dagger)}(\boldsymbol{r},t) = \left[\Psi_H^{(\dagger)}(\boldsymbol{r},t), H\right]_-$
- **Result**: Martin-Schwinger Hierarchy for 1-, 2-, ..., s-particle NEGF

$$\{\mathrm{i}\,\partial_t - H_1(\boldsymbol{r},t)\}\,G(1,1') = \delta_{\mathcal{C}}(1-1') \pm \mathrm{i}\int \mathrm{d}2\,H_2(1-2)G(1,2,1,2^+)$$

$+$ adjoint equation

with $H_2(1-1') = \delta_{\mathcal{C}}(t-t')\,H_2(\boldsymbol{r}-\boldsymbol{r}')$, $\delta_{\mathcal{C}}(1-1') = \delta_{\mathcal{C}}(t-t')\,\delta(\boldsymbol{r}-\boldsymbol{r}')$ and 2^+ refers to $t \to t_2 + \varepsilon_{>0}$

- Formal decoupling of the hierarchy introducing **self-energy** Σ

$$\pm\mathrm{i}\int \mathrm{d}2\,H_2(1-2)G(1,2,1,2^+) = \int \mathrm{d}2\,\Sigma(1,2)G(2,1')$$

- **Conserving approximations**[1]

$$\Sigma(1,1')[G] = \frac{\delta\Phi}{\delta G(1,1')}$$

[1] G. Baym, Phys. Rev. **127**, 1391 (1962)

Equations of motion for Keldysh Green's functions

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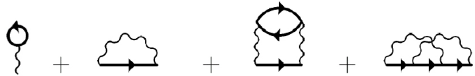
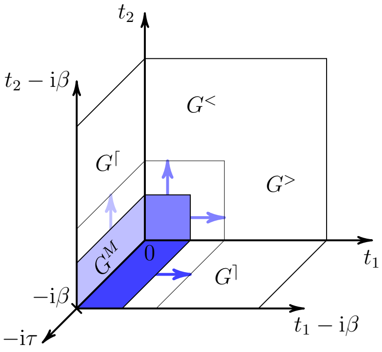
- **Example** for Σ: 1st and 2nd order diagrams $\Rightarrow$ Hartree-Fock $+$ second Born



[1] G. Baym, Phys. Rev. **127**, 1391 (1962)

Numerical solution of Keldysh-Kadanoff-Baym equations[1]

- Full two-time solutions: Danielewicz, Schäfer, Köhler/Kwong, Bonitz/Semkat, Haug, Jahnke, van Leeuwen, Balzer ...
- $G(\cdot, t_1 + \mathrm{i}\tau_1; \cdot, t_2 + \mathrm{i}\tau_2)$



- Initial conditions after the equilibrium problem is solved (Dyson equation $\rightarrow G^M(\tau)$)

$$G^\lceil(0, -\mathrm{i}\tau) = \mathrm{i}G^M(\tau)$$
$$G^\rceil(-\mathrm{i}\tau, 0) = \mathrm{i}G^M(-\tau)$$
$$G^M(-\tau + \beta) = -G^M(\tau)$$
$$G^\gtrless(0, 0) = \mathrm{i}G^M(0^\pm)$$

- Parallel algorithm[2] (MPI)

1 A. Stan, N.E. Dahlen, and R. van Leeuwen, J. Chem. Phys. **130**, 224101 (2009)

2 K. Balzer etal., acccepted in Phys. Rev. A (2010)

Outline

Derivation of single-time kinetic equations

PHYSICAL REVIEW E VOLUME 60, NUMBER 4 OCTOBER 1999

Quantum kinetic theory of plasmas in strong laser fields

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(Received 9 February 1999)

A kinetic theory for quantum many-particle systems in time-dependent electromagnetic fields is developed based on a gauge-invariant formulation. The resulting kinetic equation generalizes previous results to quantum systems and includes many-body effects. It is, in particular, applicable to the interaction of strong laser fields with dense correlated plasmas. [S1063-651X(99)10609-3]

Kadanoff-Baym equations

$$\left[i\hbar \frac{\partial}{\partial t_1} - \frac{1}{2m_a}\left(\frac{\hbar}{i}\nabla_1 - \frac{e_a}{c}\mathbf{A}(1) \right)^2 - e_a\phi(1) \right] g_a^{\gtrless}(1,1')$$

$$+ \int d\bar{\mathbf{r}}_1\, \Sigma_a^{\mathrm{HF}}(1,\bar{\mathbf{r}}_1 t_1) g_a^{\gtrless}(\bar{\mathbf{r}}_1 t_1, 1')$$

$$= \int_{t_0}^{t_1} d\bar{1}\, [\Sigma_a^{>}(1,\bar{1}) - \Sigma_a^{<}(1,\bar{1})]\, g_a^{\gtrless}(\bar{1},1')$$

$$- \int_{t_0}^{t_1'} d\bar{1}\, \Sigma_a^{\gtrless}(1,\bar{1})\, [g_a^{>}(\bar{1},1') - g_a^{<}(\bar{1},1')],$$

Self-energy and spectral function

Σ in second Born approximation

$$\Sigma_a^{\gtrless}(\mathbf{k}_a;t_1,t_1')=\sum_b \int \frac{d\mathbf{k}_b d\bar{\mathbf{k}}_a d\bar{\mathbf{k}}_b}{(2\pi\hbar)^9}|V_{ab}(\mathbf{k}_a-\bar{\mathbf{k}}_a)|^2$$
$$\times(2\pi\hbar)^3\delta(\mathbf{k}_a+\mathbf{k}_b-\bar{\mathbf{k}}_a-\bar{\mathbf{k}}_b)$$
$$\times\hbar^2 g_a^{\gtrless}(\bar{\mathbf{k}}_a;t_1,t_1')g_b^{\gtrless}(\bar{\mathbf{k}}_b;t_1,t_1')$$
$$\times g_b^{\lessgtr}(\mathbf{k}_b;t_1',t_1).$$

Full field dependence included in $g^{\gtrless}$

Nonequilibrium spectral function in field

$$a_a(\mathbf{p};\tau,t)=\exp\left\{-\frac{i}{\hbar}\int_{t-\tau/2}^{t+\tau/2}dt'\left[\mathbf{p}-\frac{e_a}{c}\mathbf{A}(t')\right]^2/2m_a\right\}$$

Quantum kinetic equation for $E = E_0 \cos \Omega t$

$$\left\{\frac{\partial}{\partial t} + e_a \mathbf{E}(t) \cdot \nabla_{k_a}\right\} f_a(\mathbf{k}_a, t) = \sum_b I_{ab}(\mathbf{k}_a, t)$$

$$\begin{aligned}
I_{ab}(\mathbf{k}_a, t) = 2 \int & \frac{d\mathbf{k}_b d\bar{\mathbf{k}}_a d\bar{\mathbf{k}}_b}{(2\pi\hbar)^6} \frac{1}{\hbar^2} |V_{ab}(\mathbf{k}_a - \bar{\mathbf{k}}_a)|^2 \\
& \times \delta(\mathbf{k}_a + \mathbf{k}_b - \bar{\mathbf{k}}_a - \bar{\mathbf{k}}_b) \\
& \times \int_{t_0}^{t} d\bar{t} \operatorname{Re} \exp\left\{ \frac{i}{\hbar} [(\epsilon_{ab} - \bar{\epsilon}_{ab})(t - \bar{t}) \right. \\
& \left. - (\mathbf{k}_a - \bar{\mathbf{k}}_a) \cdot \mathbf{R}_{ab}(t, \bar{t})] \right\} \{ \bar{f}_a \bar{f}_b [1 - f_a][1 - f_b] \\
& - f_a f_b [1 - \bar{f}_a][1 - \bar{f}_b] \}_{|\bar{t}}, \qquad (27)
\end{aligned}$$

$$\begin{aligned}
\mathbf{R}_{ab}(t, \bar{t}) = \left(\frac{e_a}{m_a} - \frac{e_b}{m_b}\right) & \left[\frac{\mathbf{E}_0 \cdot (t - \bar{t})}{\Omega} \sin \Omega t \right. \\
& \left. + \frac{\mathbf{E}_0}{\Omega^2} (\cos \Omega t - \cos \Omega \bar{t}) \right].
\end{aligned}$$

High frequency limit

$$I_{ab}(\mathbf{k}_a, t) = 2\int \frac{d\mathbf{k}_b d\bar{\mathbf{k}}_a d\bar{\mathbf{k}}_b}{(2\pi\hbar)^6} \frac{1}{\hbar^2} |V_{ab}(\mathbf{k}_a - \bar{\mathbf{k}}_a)|^2$$

$$\times \delta(\mathbf{k}_a + \mathbf{k}_b - \bar{\mathbf{k}}_a - \bar{\mathbf{k}}_b)$$

$$\times \sum_n J_n^2\left(\frac{\mathbf{q}\cdot\mathbf{w}_{ab}^0}{\hbar\Omega}\right) \int_{t_0}^{t} d\bar{t} \cos\left[\frac{1}{\hbar}(\epsilon_{ab} - \bar{\epsilon}_{ab}\right.$$

$$\left. - \mathbf{q}\cdot\mathbf{w}_{ab}(t) + n\hbar\Omega)(t-\bar{t})\right] \{\bar{f}_a \bar{f}_b [1 - f_a]$$

$$\times [1 - f_b] - f_a f_b [1 - \bar{f}_a][1 - \bar{f}_b]\}_{|\bar{t}}.$$

where

$$\boldsymbol{w}_{ab}(t) = \frac{E_0}{\Omega}\left[\frac{e_a}{m_a} - \frac{e_b}{m_b}\right] \sin \Omega t$$

Laser plasmas: Numerical results

PHYSICAL REVIEW E, VOLUME 64, 026405

Harmonics generation in electron-ion collisions in a short laser pulse

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(Received 30 March 2001; published 18 July 2001)

Anomalously high generation efficiency of coherent higher field harmonics in collisions between *oppositely charged particles* in the field of femtosecond lasers is predicted. This is based on rigorous numerical solutions of a quantum kinetic equation for dense laser plasmas that overcomes limitations of previous investigations.

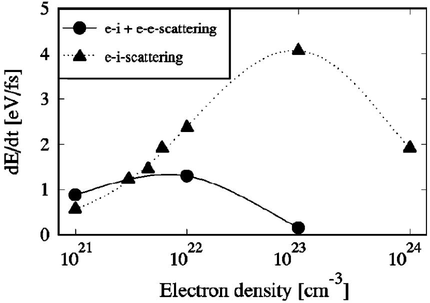


FIG. 1. Average electron kinetic energy increase per time versus density, with and without *e-e* scattering included. Field amplitude and wavelength are $E_0 = 3\times10^8$V/cm and $\lambda = 500$ nm. The initial plasma temperature is $T_0 = 20\,000$ K.

Harmonics in nonequilibrium laser plasma

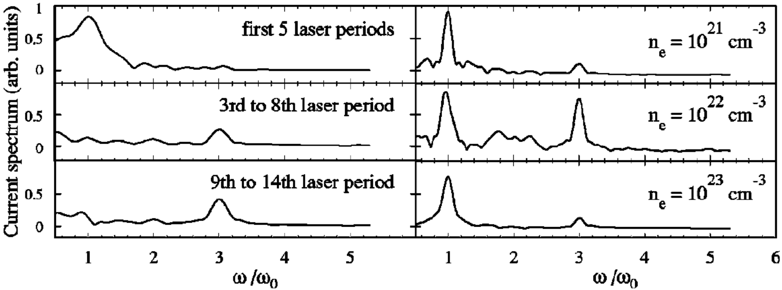


FIG. 2. Electrical current spectrum in a time-dependent electric field. The higher harmonics are the sole result of electron-ion collisions. Left column—time evolution of the spectrum for $n = 10^{22}$ cm^{-3}, from averaging over five laser periods at different moments (see text in this figure). Right column—spectrum (from averaging over the full calculation) for various densities. Other parameters are the same as in Fig. 1.

Dynamics of electron distribution function

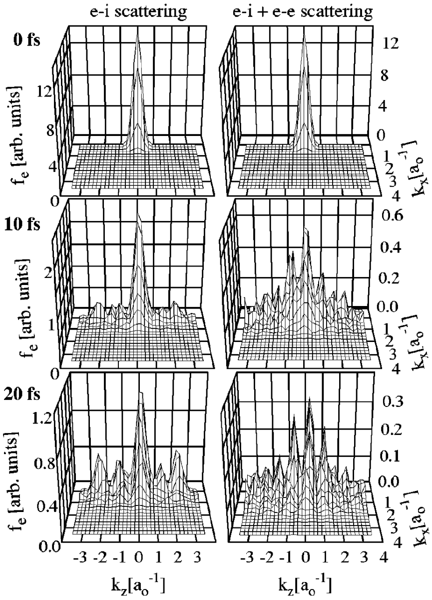
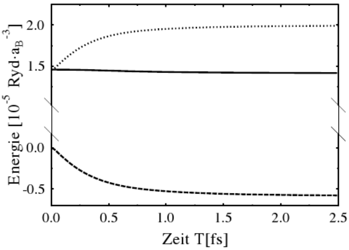


FIG. 4. Evolution of the electron distribution function in momentum space (the field is in the k_z direction) with only electron-ion collisions included (left column) and with electron-ion *and* electron-electron collisions (right column), respectively. Figure shows snapshots at $t=0$ (upper figures), after 6 laser periods (middle) and after 12 laser periods (bottom), when the main peak of f_e is at the origin (the distribution as a whole oscillates with the field in k_z direction and is isotropic in the k_x-k_y plane). Note the different vertical scales.

Sub-femtosecond energy relaxation in dense plasma[1]

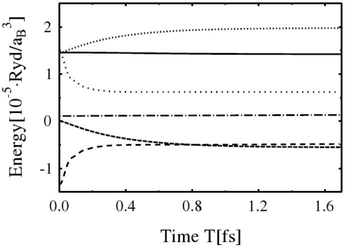
- **Dense hydrogen plasma**, $T = 10000$ K , $n = 10^{21}$ cm^{-3} , $k = 0.6/a_B$
- Solution of KKBE conserves total energy $\langle H\rangle(t) = \langle T\rangle(t) + \langle U\rangle(t) = \langle H\rangle(0)$

Initial state uncorrelated
(zero correlation energy $\langle U\rangle$)



- Correlations build up $\rightarrow$ Increase of $|\langle U\rangle| \rightarrow$ Increase of kinetic energy $\langle T\rangle$
- $\langle T\rangle$ and $\langle U\rangle$ saturate at correlation time $t \approx \tau_{\rm corr} = \omega_{\rm pl}^{-1}$

Uncorrelated vs. over-correlated initial state



- Preparing system in over-correlated initial state leads to cooling

[1] D. Semkat, D. Kremp, and M. Bonitz, Phys. Rev. E **59**, 1557 (1999)

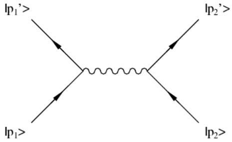
Limitations of Boltzmann-type kinetic equations

$$\left\{\frac{\partial}{\partial t}+\frac{\partial E}{\partial \mathbf{p}}\frac{\partial}{\partial \mathbf{R}}-\frac{\partial E}{\partial \mathbf{R}}\frac{\partial}{\partial \mathbf{p}}\right\} f(\mathbf{p},\mathbf{R},t)=I(\mathbf{p},\mathbf{R},t) \qquad (1)$$

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

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

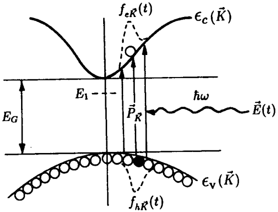
- Example: Quantum Lenard-Balescu collision integral (Coulomb scattering)



- Properties of Equation (1):
  1. Conservation of kinetic energy, $\frac{d}{dt}\langle E\rangle(t)=0$
  2. Equilibrium solution: $f(p,t)\rightarrow$ Bose/Fermi/Maxwell distribution $\rightarrow$ thermodynamic and transport properties of ideal gas
  3. Eq. (1) limited to times larger than correlation time, $t\gg\tau_{corr}$

Properties (1)–(3) in conflict with present goal ⇒ Generalizations necessary

Optical excitation of semiconductors



N. H. KWONG et al.: Kadanoff-Baym Equation Results for Optically Excited E–H Plasmas 197

phys. stat. sol. (b) **206**, 197 (1998)

Subject classification: 78.47.+p; 71.35.Ee; 78.66.Fd; S7.12

Semiconductor Kadanoff-Baym Equation Results for Optically Excited Electron–Hole Plasmas in Quantum Wells

N. H. KWONG (a), M. BONITZ (a), R. BINDER (b), and H. S. KÖHLER (c)

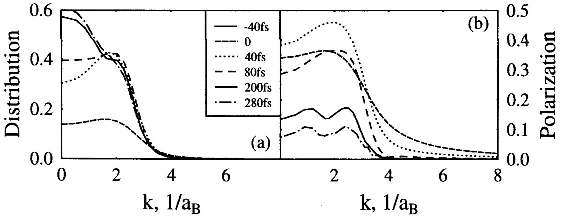
We present results from solutions of the semiconductor Kadanoff-Baym equations (full two-time semiconductor Bloch equations) with self-energies in quasistatic Born approximation, for GaAs single quantum wells. We concentrate on memory and correlation effects under fs-pulse excitation conditions. A remarkable feature is the observed kinetic energy increase which is due to the build-up of correlations among the generated carriers. We demonstrate that the two-time approach is (i) very well suited to study correlation phenomena both on short and long times, thereby avoiding well-known problems of one-time kinetic equations, and (ii) that it is becoming practical with the use of efficient integration techniques.

$$\left[i\hbar\,\frac{\partial}{\partial t_1} - \varepsilon_{\mu_1}(\mathbf{k})\right] G^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2) = \sum_{\bar\mu} \hbar\Omega_{\mu_1\bar\mu}(\mathbf{k}t_1)\, G^{\gtrless}_{\bar\mu\mu_2}(\mathbf{k}t_1t_2) + I^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2)\,,$$

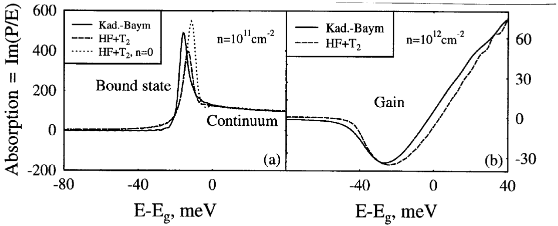
$$\left[-i\hbar\,\frac{\partial}{\partial t_2} - \varepsilon_{\mu_2}(\mathbf{k})\right] G^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2) = \sum_{\bar\mu} G^{\gtrless}_{\mu_1\bar\mu}(\mathbf{k}t_1t_2)\, \hbar\Omega_{\bar\mu\mu_2}(\mathbf{k}t_2) - I^{\gtrless *}_{\mu_2\mu_1}(\mathbf{k}t_2t_1)\,.$$

$$\hbar\Omega_{\mu_1\mu_2}(\mathbf{k}t) = -d_{\mu_1\mu_2}E(t)\,(1-\delta_{\mu_1\mu_2}) + i\hbar\sum_{\mathbf{k}'} G^{<}_{\mu_1\mu_2}(\mathbf{k}'tt)\,V(\mathbf{k}-\mathbf{k}')$$

Wigner function and absorption coefficient



Dynamics of Wigner function and polarization excited by 50 fs pulse,
$\hbar\omega = E_g + 10$ meV



Dielectric response including collisions

VOLUME 84, NUMBER 8 PHYSICAL REVIEW LETTERS 21 FEBRUARY 2000

Real-Time Kadanoff-Baym Approach to Plasma Oscillations in a Correlated Electron Gas

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(Received 30 August 1999)

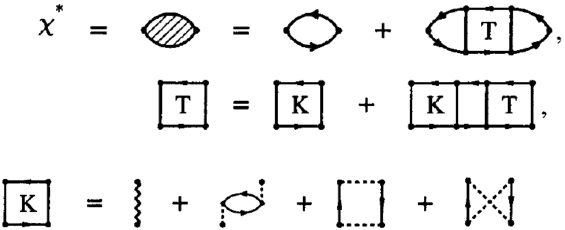
A nonequilibrium Green's functions approach to the collective response of correlated Coulomb systems at finite temperatures is presented. It is shown that solving Kadanoff-Baym-type equations of motion for the two-time correlation functions including the external perturbing field allows one to compute the plasmon spectrum with collision effects in a systematic and consistent way. The scheme has a "built-in" sum-rule preservation and is simpler to implement numerically than the equivalent equilibrium approach based on the Bethe-Salpeter equation.

$$\left(i\hbar\frac{\partial}{\partial t_1} - \epsilon_{\mathbf{k}_1}\right)G^{\gtrless}(\mathbf{k}_1 t_1; \mathbf{k}_2 t_2) = \sum_{\mathbf{q}} U(-\mathbf{q}, t_1) G^{\gtrless}(\mathbf{k}_1 - \mathbf{q}, t_1; \mathbf{k}_2 t_2) + \sum_{\bar{\mathbf{k}}} \Sigma^{\mathrm{HF}}(\mathbf{k}_1 t_1; \bar{\mathbf{k}} t_1) G^{\gtrless}(\bar{\mathbf{k}} t_1; \mathbf{k}_2 t_2) + I^{\gtrless}(\mathbf{k}_1 t_1; \mathbf{k}_2 t_2)$$

$$S(\omega, \mathbf{q}_0) = -\frac{1}{\pi n_0 U_0(\omega)} \sum_{\mathbf{k}} \mathrm{Im} G_{10}^{<}(\mathbf{k}, \omega)$$

Polarization including ladder and vertex diagrams

$$\chi^R = \chi^* + \chi^* V \chi^R$$



Correlated polarization from Σ in second Born approximation

Density response and dynamic structure factor

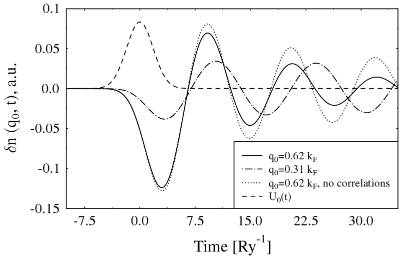


FIG. 1. Density fluctuation of a strongly correlated electron gas for two wave numbers. For comparison, the uncorrelated response for one wave number (dotted line) and the exciting field (dashes) are shown, too. k_F denotes the Fermi momentum, Ry = 13.6 eV.

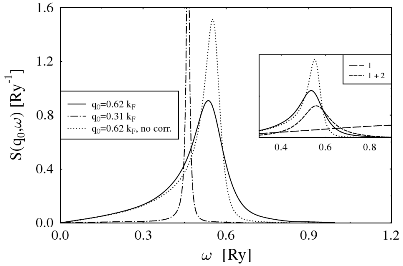


FIG. 2. Dynamic structure factor (12) for the correlated electron gas of Fig. 1 (same line styles). Inset shows S for $q_0 = 0.62k_F$ and contains two other approximations to the correlations corresponding to retaining the first diagram in Eq. (17) and first plus second diagrams, respectively.

NEGF approach to inhomogeneous systems

Consider Ne^- Hamiltonian [in a.u.]:

$$\hat{H}(t) = \hat{T} + \hat{V}(t) + \hat{U} \,,$$

$$\hat{T} = \sum_{i=1}^{N} -\frac{1}{2}\nabla_i^2 \,, \qquad \hat{V}(t) = \sum_{i=1}^{N} V(x_i, t) \,, \qquad \hat{U} = \sum_{i<j}^{N} U(|x_i - x_j|)$$

Keldysh-Kadanoff-Baym equations, $1 = (x, t, \sigma)$

$$\left\{ \mathrm{i}\partial_t - \left(-\frac{1}{2}\nabla_x^2 + V(x,t) \right) \right\} G(1,1') = \delta_{\mathcal{C}}(1-1') + \int_{\mathcal{C}} \mathrm{d}2\, \Sigma[G,U](1,2)\, G(2,1')$$

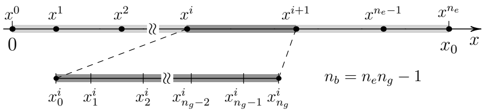
$$+ \text{ adjoint equation } \; t \leftrightarrow t'$$

Spin-polarized/restricted ansatz
(spin degeneracy $\xi \in \{1, 2\}$)

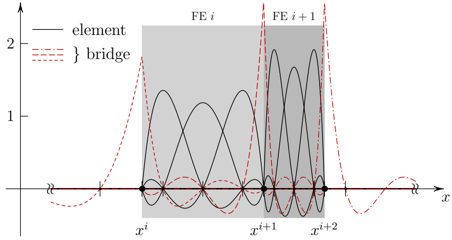
$\xi = 1$: $|\uparrow\rangle\ |\uparrow\rangle\ |\uparrow\rangle\ \ldots$
$\xi = 2$: $|\uparrow\downarrow\rangle\ |\uparrow\downarrow\rangle\ \ldots$

$$G(1,1') \rightarrow G(xt, x't') \,, \qquad \Sigma[G,U](1,1') \rightarrow \Sigma_\xi[G,U](xt, x't')$$

Finite-element DVR



i. partition $[0, x_0]$ into n_e FEs i



ii. $\forall$ FEs there exist points x_m^i and weigths w_m^i of the generalized Gauss-Lobatto quadrature

$\Rightarrow$ construct a local DVR basis $\chi_m^i(x)$

$$\chi_m^i(x) = \frac{1}{\sqrt{w_m^i}} \prod_{n \neq m} \frac{x - x_n^i}{x_m^i - x_n^i}$$

FE-DVR ansatz[1] $(n_b = n_e n_g - 1)$

$$G(xt, x't') = \sum_{im} \sum_{\bar{i}\bar{m}} \chi_m^i(x)\, \chi_{\bar{m}}^{\bar{i}}(x')\, G_{m\bar{m}}^{i\bar{i}}(t, t')\;, \qquad x, x' \in [0, x_0]$$

[1] K. Balzer, S. Bauch and M. Bonitz, Phys. Rev. A **81**, 022510 (2010)

KBE in FE-DVR representation[1]

KBEs transform into EoM for matrix $G^{i\bar{i}}_{m\bar{m}}(t,t')$ Einstein notation!

$$\left\{ \mathrm{i}\partial_t - \left(T^{i\bar{i}}_{m\bar{m}} + V^{i\bar{i}}_{m\bar{m}}(t) \right) \right\} G^{\bar{i}i'}_{\bar{m}m'}(t,t') = \delta_{\mathcal{C}}(t-t') + \int_{\mathcal{C}} \mathrm{d}\bar{t}\, \Sigma^{i\bar{i}}_{\xi,m\bar{m}}[G,U](t,\bar{t})\, G^{\bar{i}i'}_{\bar{m}m'}(\bar{t},t')$$

$+$ adjoint equation $t \leftrightarrow t'$

<u>FE-DVR matrix elements</u>:

$V^{i\bar{i}}_{m\bar{m}}(t) \propto \delta_{ii'}\delta_{mm'}$ is diagonal

$T^{i\bar{i}}_{m\bar{m}}$ is block-diagonal

computed by the generalized Gauss-Lobatto quadrature rule

Self-energy $\Sigma^{i\bar{i}}_{\xi,m\bar{m}}[G,U](t,\bar{t})$ involves matrix elements of $U(|x-x'|)$

$$U^{i_1 i_2, i_3 i_4}_{m_1 m_2, m_3 m_4} = \int_0^{x_0} \mathrm{d}x \int_0^{x_0} \mathrm{d}x'\, \chi^{i_1}_{m_1}(x)\, \chi^{i_3}_{m_3}(x')\, U(|x-x'|)\, \chi^{i_2}_{m_2}(x)\, \chi^{i_4}_{m_4}(x')$$

$$= \delta_{i_1 i_2}\delta_{i_3 i_4}\delta_{m_1 m_2}\delta_{m_3 m_4}\, \tilde{U}^{i_1 i_3}_{m_1 m_3} \quad \text{(high degree of diagonality)}$$

[1] K. Balzer, S. Bauch and M. Bonitz, Phys. Rev. A **81**, 022510 (2010)

Self-energy functionals

Self-energy in FE-DVR representation ($\xi \in \{1,2\}$)

$$\Sigma^{ii'}_{\xi,mm'}(t,t') = \delta_{\mathcal{C}}(t-t')\Sigma^{\mathrm{HF},ii'}_{\xi,mm'}(t) + \Sigma^{\mathrm{Corr},ii'}_{\xi,mm'}(t,t')$$

Hartree-Fock (HF) contribution[1]:

$\mathcal{O}(n_b) \quad \neg\mathcal{O}(n_b^2)$

$$\Sigma^{\mathrm{HF},ii'}_{\xi,mm'}(t) = -\mathrm{i}\left\{\delta_{ii'}\delta_{mm'}\xi\sum_{i_1m_1} G^{i_1i_1}_{m_1m_1}(t,t^+)\,\tilde{U}^{ii_1}_{mm_1} - G^{i'i}_{m'm}(t,t^+)\,\tilde{U}^{i'i}_{m'm}\right\}$$

Correlations in second Born (2ndB) approximation[1]:

$$\Sigma^{\mathrm{Corr},ii'}_{\xi,mm'}(t,t') = \sum_{i_1m_1}\sum_{i_2m_2}\left\{\xi\, G^{ii'}_{mm'}(t,t')\,G^{i_1i_2}_{m_1m_2}(t,t') - G^{ii_2}_{mm_2}(t,t')\,G^{i_1i'}_{m_1m'}(t,t')\right\}\times$$

$$\times\; G^{i_2i_1}_{m_2m_1}(t',t)\,\tilde{U}^{ii_2}_{mm_2}\,\tilde{U}^{i'i_1}_{m'm_1}$$

$\mathcal{O}(n_b^2)$
$\neg\mathcal{O}(n_b^6)$

[1] K. Balzer, S. Bauch and M. Bonitz, Phys. Rev. A **81**, 022510 (2010)

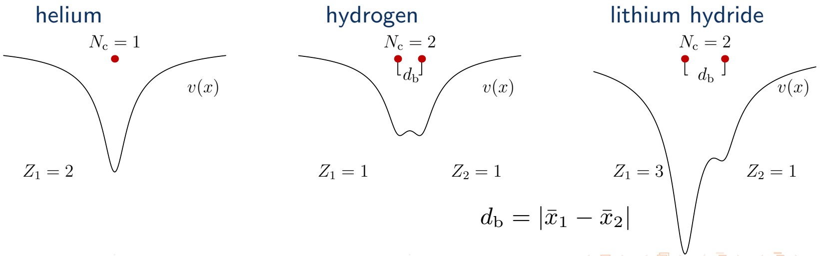
He, H_2 and LiH—One-dimensional models

Ne^--Hamiltonian (in a.u.)

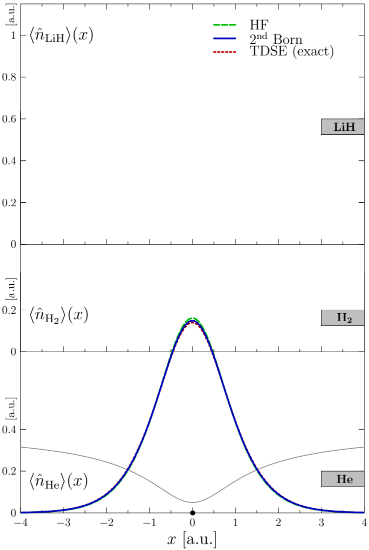
$$\hat{H}(t) = \hat{T} + \hat{V}(t) + \hat{U}$$

$$= \sum_{i=1}^{N} \left\{ -\frac{1}{2}\nabla_i^2 + \sum_{n=1}^{N_c} \frac{-Z_n}{\sqrt{(x_i - \bar{x}_n)^2 + c_n}} + E(t)x_i \right\} + \sum_{i<j} \frac{1}{\sqrt{(x_i - x_j)^2 + c}}$$

N_c: number of nuclei
Z_n: atomic number of nucleus n
$\bar{x}_n$: nuclei positions, determine molecular geometry
$c_n = c = 1 \; \forall \; n$: soft-core Coulomb potentials/interaction



$1e^-$-densities, energies and bond lengths[1]



He $(2e^-)$

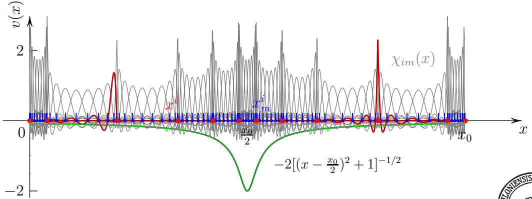
Parameters:

$x_0 = 50$ a.u.

$n_b = 153$ FE-DVR basis functions

Ground state energy E_{gs} [Ha]:

HF	-2.224210
2ndB	$-2.233419 \approx 65\%$ of corr. energy
exact	-2.238258



[1] K. Balzer, S. Bauch and M. Bonitz, Phys. Rev. A **81**, 022510 (2010)

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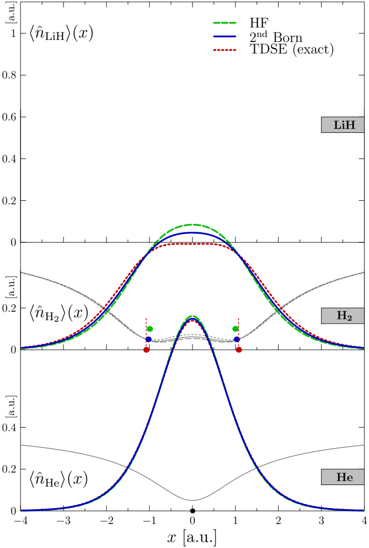


H_2 ($2e^-$) and LiH ($4e^-$)

Self-consistent results (d_b, E_b) obtained by scanning PES:

Bond-length d_b [a.u.]:

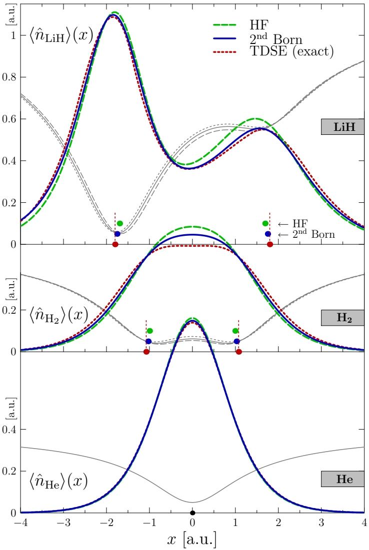
	HF	2ndB	exact
H_2	1.9925	2.0561	2.151
LiH	3.3860	3.5053	3.6 · ·

Binding energy E_b [Ha]:

	HF	2ndB	exact
H_2	−1.3531	−1.3740	−1.391
LiH	−4.8534	−4.8886	−4.91 ·

[1] K. Balzer, S. Bauch and M. Bonitz, Phys. Rev. A **81**, 022510 (2010)

$1e^-$-densities, energies and bond lengths[1]



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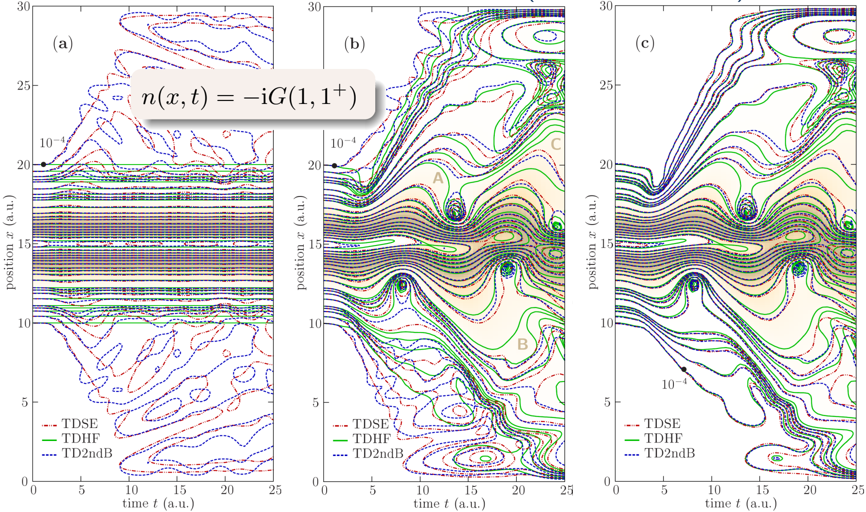
[1] K. Balzer, S. Bauch and M. Bonitz, Phys. Rev. A **81**, 022510 (2010)

1D helium—TDHF & TD2ndB vs. TDSE[1]

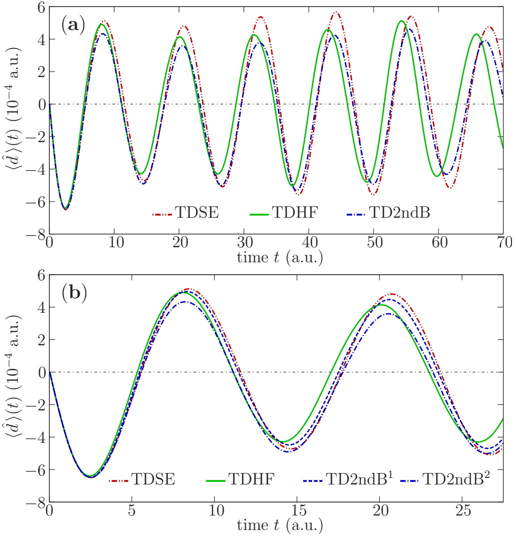
—no uv field— —uv field ($E_0 = 0.1$, $\omega = 0.54$)—



—prepared in HF ground (initial) state— —fully self-consistent—

[1] K. Balzer etal., acccepted in Phys. Rev. A (2010)

1D helium—time-dependent dipole moment[1]



Small δ-kick excitation

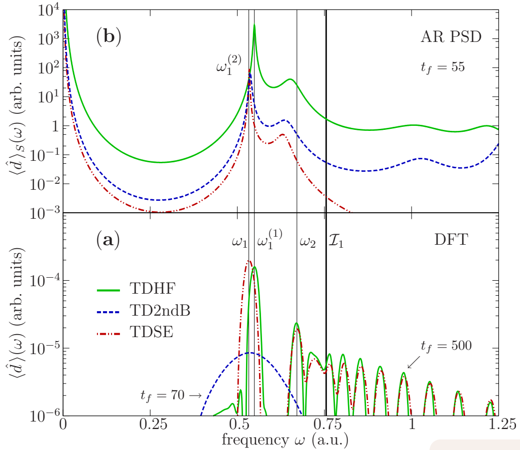
(a) TD2ndB: HF initial state

(b) TD2ndB: Comparison of HF (TD2ndB^2) and 2ndB initial state (TD2ndB^1)

Fourier transform of $\langle\hat{d}\rangle(t)$ gives absorption spectrum (dipole strength) $\langle\hat{d}\rangle(\omega)$

[1] K. Balzer etal., acccepted in Phys. Rev. A (2010)

1D helium—time-dependent dipole moment[1]



$$\omega_1 = 0.533,$$
$$\omega_1^{(1)} = 0.549,\ \omega_1^{(2)} = 0.537$$

Small δ-kick excitation

(a) TD2ndB: HF initial state

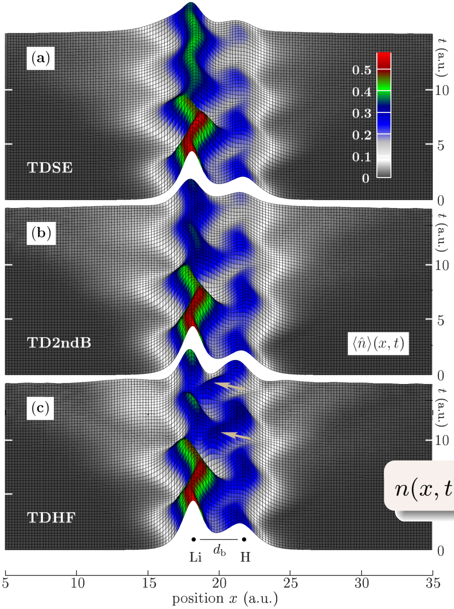
(b) TD2ndB: Comparison of HF (TD2ndB[2]) and 2ndB initial state (TD2ndB[1])

Fourier transform of $\langle \hat{d} \rangle(t)$ gives absorption spectrum (dipole strength) $\langle \hat{d} \rangle(\omega)$

(a) Discrete Fourier Transform (DFT) **(b)** Auto-regression power spectral density (AR PSD)

[1] K. Balzer etal., acccepted in Phys. Rev. A (2010)

1D LiH—TDHF & TD2ndB vs. TDSE[1]



uv excitation of LiH $(4e^-)$

Comparison of fully self-consistent calculations

Laser field parameters:

$$E_0 = 0.75$$
$$\omega = 1.5$$

$$n(x,t) = -\mathrm{i}G(1,1^+)$$

Outline

Summary

1. NEGFs allow for a systematic theoretical approach to correlated many-body systems
2. Strong fields, short pulses are naturally included nonperturbatively
3. Strict derivation of kinetic equations and generalizations
4. Direct numerical solution of the Keldysh-Kadanoff-Baym equations possible for simple systems
5. First applications to small atoms and molecules (using FE-DVR basis). Favorable scaling with N
6. Applicability to x-ray range expected

For more details see

www.theo-physik.uni-kiel.de/~bonitz

network "Quantum Many-Body Dynamics out of Equilibrium"
www.theo-physik.uni-kiel.de/~bonitz/kbet2.thml

References

Further publications:

S. Bauch, K. Balzer, and M. Bonitz, *Electronic correlations in double ionization of atoms in pump-probe experiments*, arXiv:1005.1253, accepted for publication in Euro. Phys. Lett. (2010)

S. Bauch, and M. Bonitz, *Fast electron generation by Coulomb scattering on spatially correlated ions in a strong laser field*, arXiv:0904.4855, Contrib. Plasma Phys. 49, 558-567 (2009)

S. Bauch, and M. Bonitz, *Angular distributions of atomic photoelectrons produced in the UV and XUV regimes*, arXiv:0806.4501, Phys. Rev. A 78, 043403 (2008)

Books:

M. Bonitz, *Quantum Kinetic Theory* (Teubner, Stuttgart/Leipzig 1998)

M. Bonitz, and D. Semkat (Eds.), *Introduction to Computational Methods in Many-Body Physics* (Rinton Press, Princeton, 2006)

Reviews:

Progress in Nonequilibrium Green's functions I-IV