Quantum kinetic equations
Lecture #3

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December 5, 2008

Outline

Basic numerical concepts I

Question of representation:

(a) Homogeneous systems (e.g. plasmas, correlated electron gases)

$$G(\mathbf{r}_1 t_1, \mathbf{r}_{\bar{1}} t_{\bar{1}}) \longrightarrow G(\mathbf{p}; t_1, t_{\bar{1}}) \quad \text{or} \quad G(\mathbf{k}; t_1, t_{\bar{1}})$$

- G depends on a single momentum or wave vector
  $\Rightarrow$ Numerically, solve KBE on a **p**- or **k**-grid

(b) Inhomogeneous finite systems (e.g. particles in traps, quantum dots, atoms)

$$G(\mathbf{r}_1 t_1, \mathbf{r}_{\bar{1}} t_{\bar{1}}) \longrightarrow g_{mn}(t_1, t_{\bar{1}})$$

Definition:

$$G(r_1 t_1, \mathbf{r}_{\bar{1}} t_{\bar{1}}) = \sum_{m,n=0}^{n_b-1} \phi_m^*(\mathbf{r}_1)\, \phi_n(\mathbf{r}_{\bar{1}})\, g_{mn}(t_1, t_{\bar{1}})$$

- $\{\phi_m(\mathbf{r})\}$, $m = 0, 1, \ldots, n_b - 1$, denotes a complete orthonormal set of (effective) one-particle states
- Solve KBE in basis representation: Each element $G^{\gtrless, M, \rceil/\lceil}$ of G becomes a $n_b \times n_b$-matrix

Basic numerical concepts II

Initial/boundary conditions:

Either

- Build up of correlations or specify initial correlation contributions including an initial self-energy $\longrightarrow \Sigma^{\mathrm{IN}}(1,\bar{1})$, see Ref. 1

or

- Apply Kubo-Martin-Schwinger (KMS) boundary conditions to start from a equilibrium Green's function, which on the same approximate level (as its nonequilibrium counterpart) accounts for the amount of correlations initially in the system.

Most complete and systematical picture: Consider the boundary problem $\rightarrow$ starting from a self-consistent equilibrium Green's function $G^M(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; \tau)$.

1. Apply equilibrium theory: Solve Dyson's equation for $G^M(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; \tau)$
2. Extension to nonequilibrium situations: Propagate the initial state characterized by $G^M(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; \tau)$ in time according to the two-time KBE

Equilibrium—Dyson equation

1. A closed equation for $G^M(1,\bar{1})$ is obtained by taking the difference of both KBEs, evaluating all Green's functions at $1 = t_0 - i\tau_1$ and $\bar{1} = t_0 - i\tau_{\bar{1}}$

Dyson equation: $\tau \in [-\beta, +\beta]$

$$
\begin{aligned}
&\left[-\partial_\tau - H^1(\mathbf{r}_1)\right] G^M(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; \tau) \\
=\quad & \delta(\tau) + \int \mathrm{d}^3\bar{r} \int_0^\beta \mathrm{d}\bar{\tau}\; \Sigma^M\left[G^M\right](\mathbf{r}_1, \bar{\mathbf{r}}; \tau - \bar{\tau})\, G^M(\bar{\mathbf{r}}, \mathbf{r}_{\bar{1}}; \bar{\tau})\ .
\end{aligned}
$$

- $G^M(1,\bar{1})$ and $\Sigma^M(1,\bar{1})$ only depend on the time-difference $\tau = \tau_1 - \tau_{\bar{1}}$ $\Rightarrow$ consider the quantities

$$X^M(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; \tau) = -i\, X^M(\mathbf{r}_1 t_0 - i\tau_1, \mathbf{r}_{\bar{1}} t_0 - i\tau_{\bar{1}})\ , \qquad X = G, \Sigma$$

- need self-consistent solution for a given self-energy approximation
- KMS boundary conditions for τ-arguments: $G(\tau) = \pm G(\tau - \beta)$

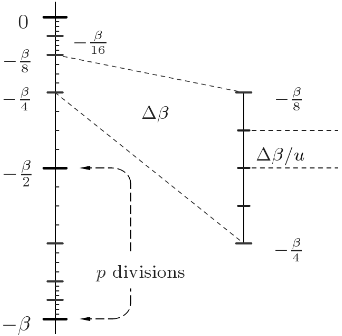
Dyson equation: Time-discretization

- Without interaction ($\Sigma \equiv 0$) the Dyson equation reads

$$\left[-\partial_\tau - H^1(\mathbf{r}_1)\right] G^M(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; \tau) = \delta(\tau)$$

  Thus, typical solution for $G(\tau)$ has exponential-like character being peaked around $\tau = 0, \pm\beta$

- Due to KMS conditions restrict solution to interval $[-\beta, 0]$. Compare with one-particle density matrix: $\rho(\mathbf{r}_1, \mathbf{r}_{\bar{1}}) = G(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; 0^-)$.



- Apply uniform power mesh (UPM) which is dense around the end-points $\tau = -\beta, 0$
- p: # of 'power' devisions
  u: # of equidistant subdevisions
  1. total # of mesh points: $M = 2up + 1$
  2. largest mesh-spacing $\Delta\tau_{\mathrm{max}} = \beta/(4u)$
  3. smallest mesh-spacing $\Delta\tau_{\mathrm{min}} = \frac{1}{2}\beta/(2^{p-1}u)$

Numerical scheme—Dyson equation

- Expand $G(1,\bar{1})$ in terms of precomputed Hartree-Fock (HF) orbitals $\phi^0_m(\mathbf{r})$, $m = 0, 1, \ldots, n_b - 1$, with eigenenergies ϵ^0_m and a chemical potential μ^0. HF Green's function with self-energy Σ^0_{ij}:

$$G^0(\mathbf{r}_1, \mathbf{r}_{\bar{1}}; \tau) = \sum_{m,n} \phi^{0\,*}_m(\mathbf{r}_1)\, \phi^0_n(\mathbf{r}_{\bar{1}})\, g^0_{mn}(\tau)\ , \quad g^0_{mn} = \delta_{mn} \frac{\exp(-\tau[\epsilon^0_i - \mu^0])}{\exp(\beta[\epsilon^0_i - \mu^0]) + 1}$$

- Dyson equation in integral form $\Leftrightarrow$ Re-interpretation as a set of n_b independent (but typically large-scale) linear systems of the form

$$\mathcal{A}\,\mathcal{X}^{(j)} = \mathcal{B}^{(j)}\ , \qquad (1)$$

with $(\mathcal{X}^{(j)})_{ip} = g^M_{ij}(\tau_p)$, $(\mathcal{B}^{(j)})_{ip} = g^0_{ij}(\tau_p)$ and coefficient matrix $(\mathcal{A})_{ip,jq} = \alpha_{ij}(\tau_p, \tau_q)$ is defined by the convolution integral

$$\alpha_{ij}(\tau, \bar{\tau}) = \delta_{ij}\delta(\tau - \bar{\tau}) - \sum_{k=0}^{n_b-1} \int_0^\beta \mathrm{d}\bar{\bar{\tau}}\, g^0_{ik}(\tau - \bar{\bar{\tau}})\, \Sigma^r_{kj}(\bar{\bar{\tau}} - \bar{\tau})$$

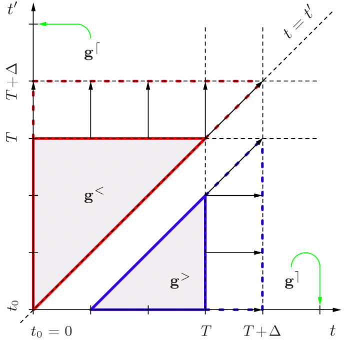
$$\Sigma^r_{ij}[g^M](\tau) = \Sigma^M_{ij}[g^M](\tau) - \delta(\tau)\, \Sigma^0_{ij}[g^0(0^-)]$$

- Eq. (1) has to be iterated! But allows for a numerically fast and accurate implementation $\longrightarrow$ Result: self-consistent correlated G^M

Real-time propagation of elements $G^{\gtrless}$ and $G^{\rceil/\lceil}$

2. For the time-propagation, we consider (without loss of generality) $t_0 = 0$.
Initial conditions[1]:

$$\mathbf{g}^<(0,0) = i\,\mathbf{g}^M(0^-)\,, \qquad \mathbf{g}^>(0,0) = i\,\mathbf{g}^M(0^+)\,,$$
$$\mathbf{g}^\rceil(0,-i\tau) = i\,\mathbf{g}^M(-\tau)\,, \qquad \mathbf{g}^\lceil(-i\tau,0) = i\,\mathbf{g}^M(\tau)\,.$$



Time-stepping procedure: more

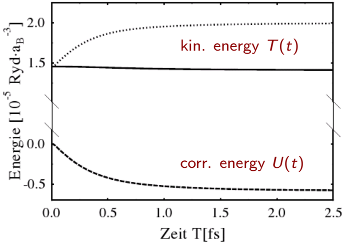
- Due to symmetry properties $[\mathbf{g}^{\gtrless}(t,t')]^\dagger = -\mathbf{g}^{\gtrless}(t',t)$ and $\mathbf{g}^>(t,t) = -i + \mathbf{g}^<(t,t)$ on the time-diagonal, restrict to propagate $\mathbf{g}^<$ on the red triangle and $\mathbf{g}^>$ on the blue triangle
- Propagate $\mathbf{g}^{\rceil/\lceil}$ on time-axes t, t'
- Each step requires to update r.h.s. of KBE (collision integrals $I^{\gtrless}(t,t')$ and $I^{\rceil/\lceil}(t,t')$ to be performed over the expanding square $[0,T]\times[0,T]$) more

[1] use matrix notation $(\mathbf{g}^M)_{ij} = g^M_{ij}$

Sub-femtosecond energy relaxation in dense plasma

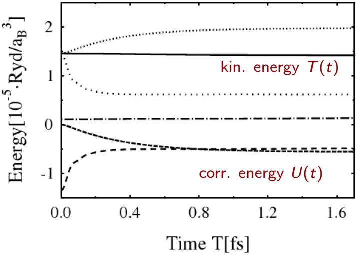
Dense hydrogen plasma: $T = 10000$ K, $n = 10^{21}$ cm^{-3}, $k = 0.6/a_B$

- Solution of the KBE equations conserves total energy $E(t) = T(t) + U(t) = E(t_0)$
- Initial state uncorrelated: zero correlation energy $U \rightarrow$ correlations build up $\rightarrow$ increase of $|U| \rightarrow$ increase of kinetic energy T



$\Rightarrow$ $T(t)$ and $U(t)$ saturate at correlation time $t \approx \tau_{\mathrm{corr}} \sim \omega_{pl}^{-1}$

- Uncorrelated vs. over-correlated initial state



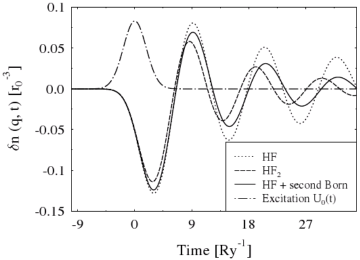
$\Rightarrow$ Preparing the system in an over-correlated initial state leads to cooling

[D. Semkat et al, Phys. Rev. E **59**, 1557 (1999)]

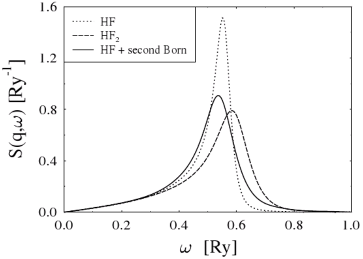
Plasmon spectrum of the correlated electron gas

Solution of the KBE for short monochromatic excitation $U(t) = U_0(t)\cos(q_0 r)$

Periodic density fluctuation, with Landau plus correlation damping



Fourier transform yields dynamic structure factor $S(q, \omega)$



- Conservation properties of the KBE guarantee exact sum rule preservation of plasmon spectrum $S(q, \omega)$
- Simple approximations for the self-energy (such as 2nd Born approximation) give high-level correlation effects, including vertex corrections, in $S(q, \omega)$

[Kwong/Bonitz, Phys. Rev. Lett. **84**, 1768 (2000)]

Application of NEGFs to quantum dot (QD) systems

***N*-electron quantum dot**:

$$\hat{H}_e = \sum_{i=1}^{N} \left(-\frac{\hbar^2}{2m_e^*} \nabla_i^2 + \frac{m_e^*}{2} \omega_0^2 \mathbf{r}_i^2 \right) + \sum_{i<j}^{N} \frac{e^2}{4\pi\varepsilon\, r_{ij}} \,, \qquad (2)$$

- Using the replacement rules $\{\mathbf{r}_i \longrightarrow \mathbf{r}_i / l_0^*,\ E \longrightarrow E/E_0^*\}$, Hamiltonian (2) transform to dimensionless form:

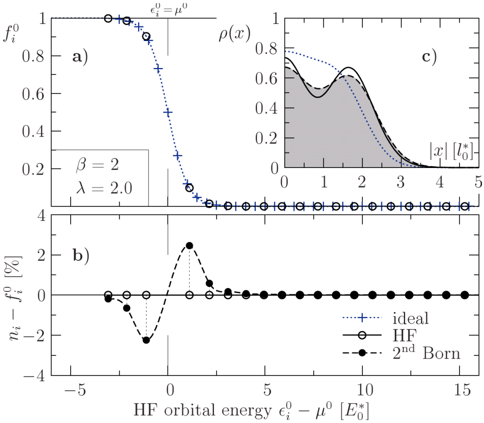
$$\hat{H}_\lambda = \frac{1}{2} \sum_{i=1}^{N} (-\nabla_i^2 + \mathbf{r}_i^2) + \sum_{i<j}^{N} \frac{\lambda}{r_{ij}} \,, \qquad \lambda = \frac{E_C}{E_0^*} = \frac{e^2}{4\pi\varepsilon\, l_0^* \hbar\omega_0} = \frac{l_0^*}{a_B}$$

$$l_0^* = \sqrt{\frac{\hbar}{m_e^* \omega_0}} \,, \qquad E_0^* = \hbar\omega_0 \,, \qquad E_C = \frac{e^2}{4\pi\varepsilon l_0^*}$$

- λ is coupling parameter (tunable via confinement frequency ω_0) $\longrightarrow$ measures the relative e^--e^- interaction strength.
- m_e^*: effective electron mass in the environement of dielectric constant ϵ, l_0^*: characteristic single-electron extension in the QD, a_B: effective electron Bohr radius, E_0^*: oscillator energy, E_C: Coulomb energy

Example: Strongly correlated QD

$N = 3$ electrons (1D parabolic confinement): $\lambda = 2$ and $\beta = 2$



c) $\rho(x)$: one-electron density

$\longrightarrow$ ideal (dotted), $\lambda = 0$

$\longrightarrow$ Hartree-Fock (solid)

$\longrightarrow$ second Born (dashed)

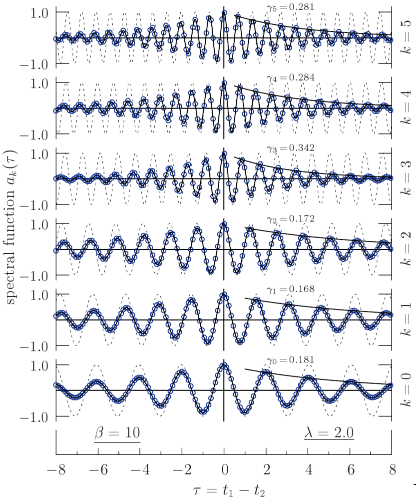
Correlations lead to weakening of density modulations

a) $f_i^0 = g_{ii}^0(0^-)$: HF (Fermi-Dirac) distribution function

b) $n_i - f_i^0 = g_{ii}^M(0^-) - g_{ii}^0(0^-)$: deviation of the second Born result from the HF distribution function in %

One-particle spectral function $a_k(\omega)$

Spectral function: Obtained from KBE time-evolution without external disturbance



- Orbital-resolved carrier spectral function[a]

$$\mathbf{a}(\tau) = i\{\mathbf{g}^{>}(T-\tau, T+\tau) - \mathbf{g}^{<}(T-\tau, T+\tau)\}$$

- Identify the diagonal (offdiagonal) elements of matrix $\mathbf{a}(\tau)$ with the intraband (interband) spectral functions

⇐ $a_k(\tau) = a_{kk}(\tau)$ for three electrons in a 1D quantum dot

Inverse hyperbolic cosine model (IHC)

$$a_k(\tau) = e^{i\omega_k\tau} \frac{1}{\cosh^{\eta_k}(\nu_k\tau)}$$

with fit parameters $\{\omega_k, \eta_k, \nu_k\}$, damping $\gamma_k = \eta_k\nu_k$

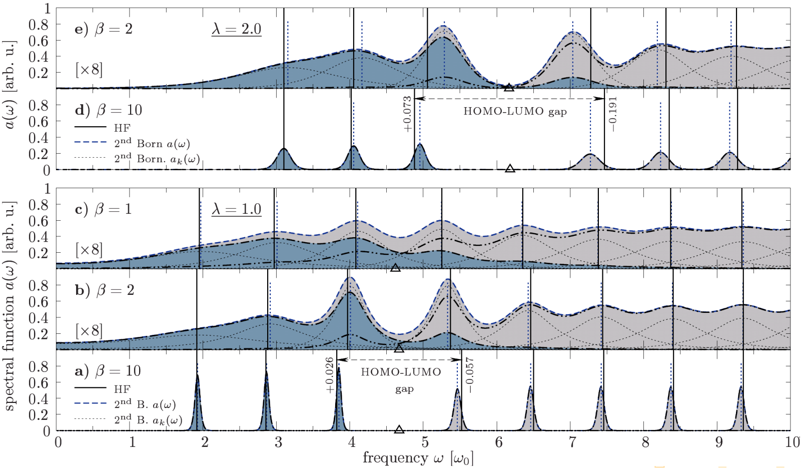
[H. Haug, L. Banyai, Solid State Comm. **100**, 303 (1998)]

[a] matrix notation $(\mathbf{a}(\tau))_{mn} = a_{mn}(\tau)$

Correlated one-particle spectral function $a(\omega) = \sum_k a_k(\omega)$

- Fourier transform of $a_k(\tau)$ → single peaks $a(\omega)$ (black dotted lines), sum of all $a_k(\omega)$ gives whole spectral function (blue dashed lines filled gray), discrete HF levels (vertical solid lines)

3 electrons in a 1D quantum dot



Correlated dipole excitation $V(\mathbf{r},t) = V_0\mathbf{r}\exp(-(t-t)^2/\Delta t^2)\cos(\omega t)$

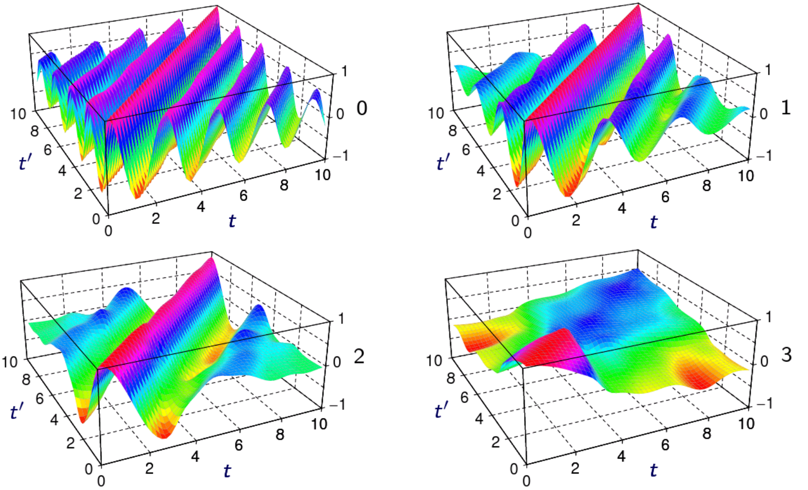


Figure: 4 e$^-$ in a 1D trap—$\lambda = 1$ and $\beta = 3$: Evolution of $\Im\mathfrak{m}\, g_{ii}^{<}(t_1, t_2)$ in the external field $V(\mathbf{r}, t)$ centered at $t_{1,2} = 4$, levels $i = 0$ to $i = 3$.

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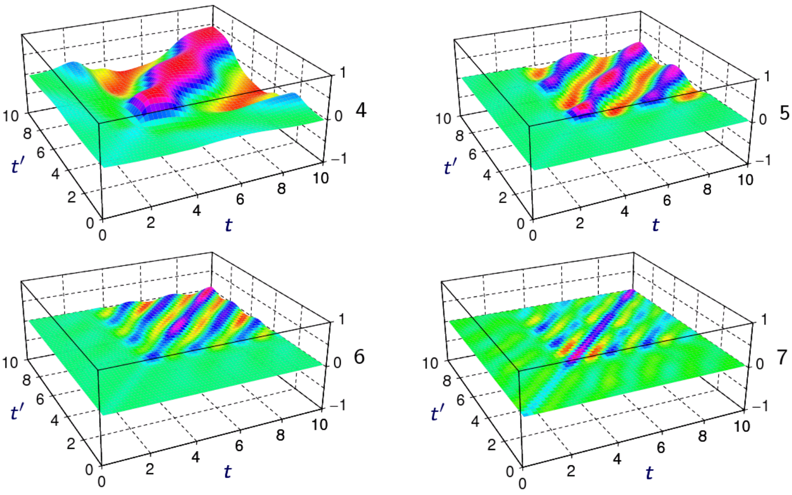

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Ionization potentials (IP) of atoms and molecules

[N.E. Dahlen and R. van Leeuwen *J. Chem. Phys.* **122**, 164102 (2005)]

	Second Born	HF	Experiment
He	24.53	24.98	24.59
Be	8.52	8.39	9.32
Ne	20.17	23.14	21.56
Mg	6.93	6.88	7.65
H_2	16.11	16.18	15.43
LiH	7.84	8.20	7.90

Table: Ground state Matsubara Green's function in second Born approx., IPs (in eV) are calculated using the Extended Koopmans Theorem (EKT).

- Form $N-1$ optimized states as a linear combination of the states formed by annihilation of one electron from the ground state: $|\phi_i^{N-1}\rangle = \int dx \;\; u_i(x)\hat{\psi}(x)|\phi_0^N\rangle$ where $|\phi_i^{N-1}\rangle$ is normalized and $u_i(x)$ is determined by the condition

$$\frac{\delta E}{\delta u_i} = 0 \; , \qquad E^{N-1}[u_i] = \frac{\langle \phi_i^N | \hat{H} | \phi_i^{N-1}\rangle}{\langle \phi_i^{N-1} | \phi_i^{N-1}\rangle}$$

- Eigenvalue equation $\int dx' \;\; \Delta(x,x')u_i(x') = (E_0^N - E_i^{N-1} - \mu)\int dx' \;\; \rho(x,x')u_i(x')$ where $\Delta = -\partial_\tau G^M(\tau)|_{\tau=0^-}$ and the density matrix is given by $\rho = G^M(0^-)$.

Atomic spectral function in an external field

[N.E. Dahlen and R. van Leeuwen, Phys. Rev. Lett. **98**, 153004 (2007)]

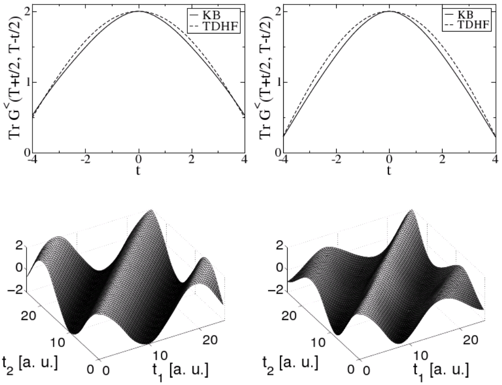

Figure: Below: Trace of $\Im m\, G^{<}(t_1, t_2)$ for the H_2 molecule (25 molecular orbitals) in its ground state (left) and in an applied electric field (right): $E(t) = \theta(t) E_0$ where $E_0 = 0.14$ *a.u*. Above: the same for a fixed value of $T = (t_1 + t_2)/2$ compared with TDHF (1 *a.u.* $= 2.418884 \cdot 10^{-17}$s)

Time-dependent dipole moment of a Be atom

[N.E. Dahlen and R. van Leeuwen, Phys. Rev. Lett. **98**, 153004 (2007)]

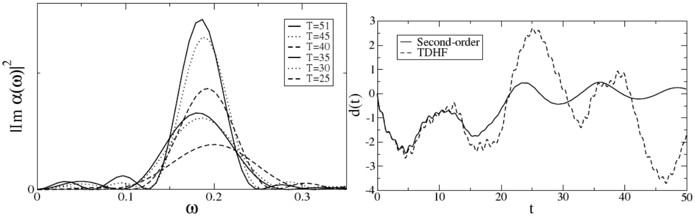

Figure: Polarizability $\alpha(\omega)$ and time dependent dipole moment $d(t)$

- $\alpha(\omega) = -1/E_0 \int_0^T \mathrm{d}t\; e^{i\omega t}\; d(t)$ with dipole moment $d(t) = -i \int \mathrm{d}^3 r\; z\, n(\mathbf{r}, t)$, propagated for various times T (using 28 basis functions).
- Left: Peak positions (for $^1\mathrm{S} \to {}^1\mathrm{P}$ excitation energy): TDHF $= 0.178$ a.u., KBE-2nd Born $= 0.189$ a.u., Experiment $= 0.194$ a.u. (1 a.u. $= 27.2$ eV)
- Right: Time dependent dipole moment calculated within HF and second Born approx., obtained with a non-perturbative "kick" $E(t) = E_0\delta(t)$, $E_0 = 1.0\ a.u.$

Conclusion

- (Ultra-)fast dynamics of Coulomb correlations. Correlation build up ⇒ Requires non-Markovian kinetic treatment
- Real-time nonequilibrium Green's functions (Keldysh/Kadanoff-Baym)
  - → Self-consistent non-perturbative treatment of strong fields
  - → Total energy conservation, exact spin statistics
- Numerical applications: propagation of NEGF in two-time plain
  - → Semiconductors, dense plasmas, electron gas
  - → Dynamics of trapped charge carriers
  - → Atoms, molecules in strong fields

Text books, reviews:

[1] *Introduction to Computational Methods in Many Body Physics*, M. Bonitz and D. Semkat (Eds.) (2006)

[2] M. Bonitz *Quantum Kinetic Theory* (1998)

[3] *Progress in Nonequilibrium Green Functions I, II, III* (2000, 2003, 2006)

- Announcement: Interdisciplinary conference *Progress in Nonequilibrium Green Functions IV*, August 17-22 2009, Glasgow, Scotland
- Web page: www.theo-physik.uni-kiel.de/∼bonitz

Real-time KBE

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Kadanoff-Baym/Keldysh equations:

- In a given basis representation $G(\mathbf{r}t, \mathbf{r}'t') \rightarrow \mathbf{g}_{mn}(t, t')$ can be rewritten in the form

$$
\begin{aligned}
i\,\partial_t\,\mathbf{g}^{>}(t,t') &= \mathbf{h}(t)\,\mathbf{g}^{>}(t,t') + \mathbf{I}_1^{>}(t,t') \\
-\,i\,\partial_t\,\mathbf{g}^{<}(t',t) &= \mathbf{g}^{<}(t',t)\,\mathbf{h}(t) + \mathbf{I}_2^{<}(t',t) \\
i\,\partial_t\,\mathbf{g}^{\rceil}(t,-i\tau) &= \mathbf{h}(t)\,\mathbf{g}^{\rceil}(t,-i\tau) + \mathbf{I}^{\rceil}(t,-i\tau) \\
-\,i\,\partial_t\,\mathbf{g}^{\lceil}(-i\tau,t) &= \mathbf{g}^{\lceil}(-i\tau,t)\,\mathbf{h}(t) + \mathbf{I}^{\lceil}(-i\tau,t)
\end{aligned}
$$

- In $\mathbf{h}(t) = \mathbf{h}^0(t) + \mathbf{\Sigma}^{\mathrm{HF}}(t)$ the Hartree-Fock self-energy and the time-dependent contribution from the single-particle Hamiltonian are collected.
- The collision integrals $I_1^{>}$, $I_2^{<}$ and $I^{\rceil/\lceil}$ are defined on the next slide!

Collision integrals

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Summary of all relevant right hand sides of the two-time KBE:

$$\mathbf{I}_1^>(t,t') = \int_0^t d\bar{t}\,\left[\boldsymbol{\Sigma}^>(t,\bar{t}) - \boldsymbol{\Sigma}^<(t,\bar{t})\right]\,\mathbf{g}^>(\bar{t},t') + \int_0^{t'} d\bar{t}\,\boldsymbol{\Sigma}^>(t,\bar{t})\,\left[\mathbf{g}^<(\bar{t},t') - \mathbf{g}^>(\bar{t},t')\right]$$
$$-i\int_0^\beta d\bar{\tau}\,\boldsymbol{\Sigma}^\rceil(t,-i\bar{\tau})\,\mathbf{g}^\lceil(-i\bar{\tau},t')$$

$$\mathbf{I}_2^<(t',t) = \int_0^{t'} d\bar{t}\,\left[\mathbf{g}^>(t',\bar{t}) - \mathbf{g}^<(t',\bar{t})\right]\,\boldsymbol{\Sigma}^<(\bar{t},t) + \int_0^t d\bar{t}\,\mathbf{g}^<(t',\bar{t})\,\left[\boldsymbol{\Sigma}^<(\bar{t},t) - \boldsymbol{\Sigma}^>(\bar{t},t)\right]$$
$$-i\int_0^\beta d\bar{\tau}\,\mathbf{g}^\rceil(t',-i\bar{\tau})\,\boldsymbol{\Sigma}^\lceil(-i\bar{\tau},t)$$

$$\mathbf{I}^\rceil(t,-i\tau) = \int_0^t d\bar{t}\,\left[\boldsymbol{\Sigma}^>(t,\bar{t}) - \boldsymbol{\Sigma}^<(t,\bar{t})\right]\,\mathbf{g}^\rceil(\bar{t},-i\tau) + \int_0^\beta d\bar{\tau}\,\boldsymbol{\Sigma}^\rceil(t,-i\bar{\tau})\,\mathbf{g}^M(\bar{\tau}-\tau)$$

$$\mathbf{I}^\lceil(-i\tau,t) = \int_0^t d\bar{t}\,\mathbf{g}^\lceil(-i\tau,\bar{t})\,\left[\boldsymbol{\Sigma}^<(\bar{t},t) - \boldsymbol{\Sigma}^>(\bar{t},t)\right] + \int_0^\beta d\bar{\tau}\,\mathbf{g}^M(\tau-\bar{\tau})\,\boldsymbol{\Sigma}^\lceil(-i\bar{\tau},t)$$