Time-dependent quantum transport using TDDFT

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Outline

- TDDFT-based approaches to transport
  - Finite systems
  - Embedding scheme
  - Master equation
- Time-dependent transport phenomena
  - Adiabatic LDA transport caluclations
  - Comparison with MBPT and tdDMRG
  - Dynamical picture of Coulomb blockade
  - Bistability
- Summary and critique of adiabatic TDDFT for transport

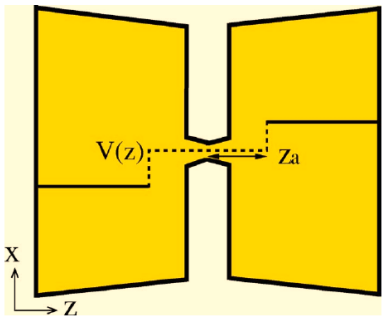
Approaches to time-dependent transport using TDDFT

TDDFT for transport: general idea

- start from contacted system L-C-R in the ground state (or in thermal equilibrium)
- at some time t_0 switch on the bias and follow time evolution
- for DC bias: expect evolution towards a steady state

Finite-system approach

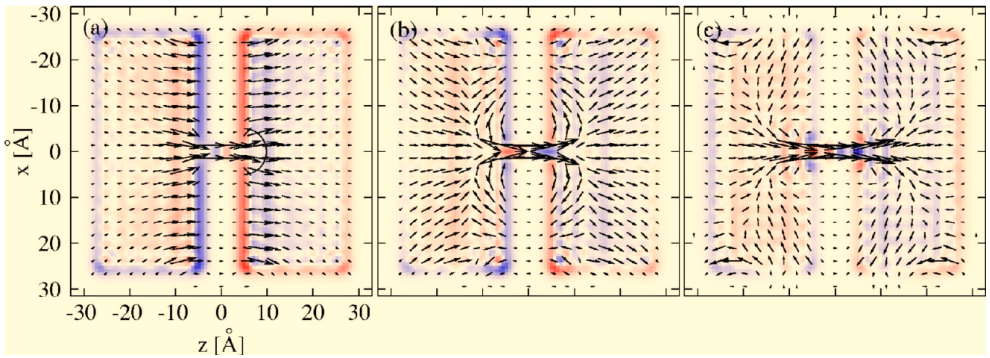
Idea: simulate nanosystem attached to large but finite leads



for $t = 0$: perform static DFT calculation with additional external potential $V(z)$ mimicking the bias

for $t > 0$: switch off $V(z)$ and perform standard KS time-evolution

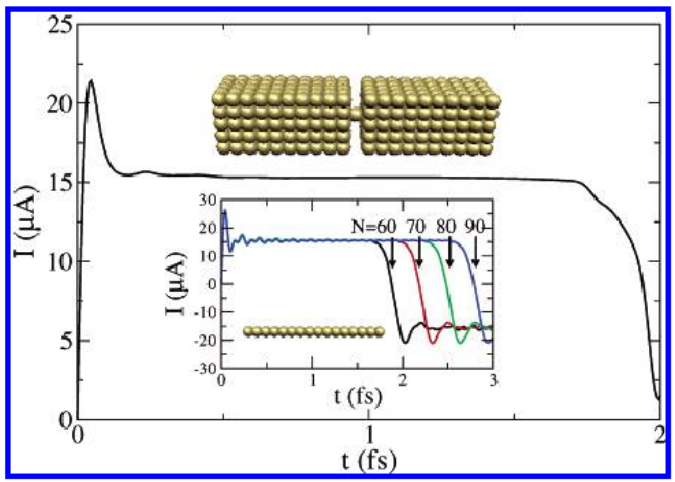
two large jellium leads connected by constriction (Sai et al, PRB **75**, 115410 (2007)): snapshots of current density in TDLDA



Finite-system approach (cont.)

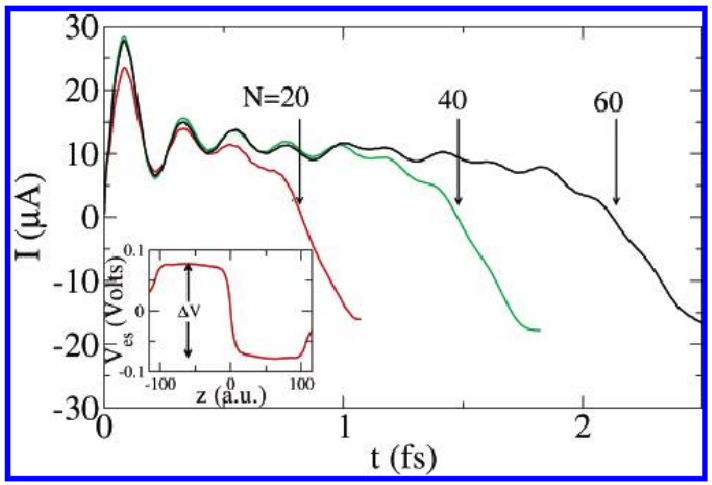
other example: tight-binding gold chain between gold electrodes (N. Bushong et al, Nano Lett. **5**, 2569 (2005))

non-interacting



clear plateau in current after transients before eventually current dies out

TDLDA



current plateau still visible but somewhat less clear

Quantum kinetic approach

Look at transport from point of view of open (electronic) system coupled to a bath (typically phonon bath)
Refs.: R. Gebauer et al, PRL **93**, 160404 (2004); Burke et al, PRL **94**, 146803 (2005)

Hamiltonian of total system

$$\hat{H}_{tot} = \hat{H}_{el} + \hat{H}_{bath} + \hat{H}_{coup}$$

reduced density operator

$$\hat{S}_{red}(t) = \mathrm{Tr}_{bath}\left[\hat{S}_{tot}(t)\right] = \mathrm{Tr}_{bath}\left[|\Psi(t)\rangle\langle\Psi(t)|\right]$$

Quantum kinetic approach

two assumption to derive eqation of motion for $\hat{S}_{red}$

- weak coupling between electrons and bath $\longrightarrow$ sufficient to go to 2nd order in $\hat{H}_{coup}$
- Markov approximation: time scale on which el. system varies is large compared to time-scale on which bath correlation functions decay

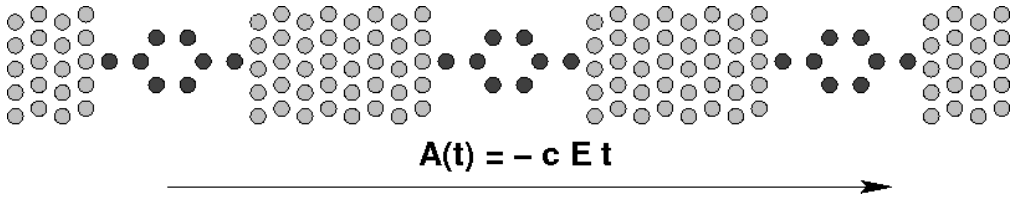
Master equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{S}_{red} = -i\left[\hat{H}_{el}, \hat{S}_{red}\right] + \breve{C}[\hat{S}_{red}]$$

with superoperator $\breve{C}[\hat{S}_{red}]$ whose explicit form depends on bath density-functionalize this approach (Burke et al (2005)): for a given superoperator, map problem of interacting electrons on an effective non-interacting one

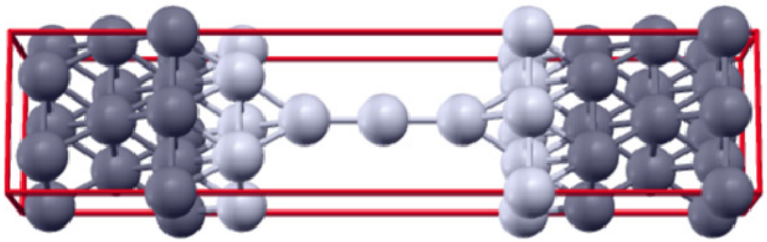
Quantum kinetic approach

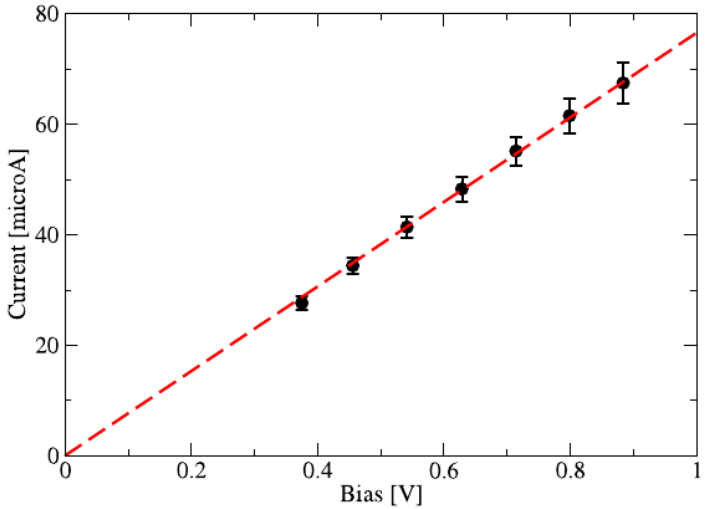
for practical convenience: use periodic boundary conditions, i.e., schematically as



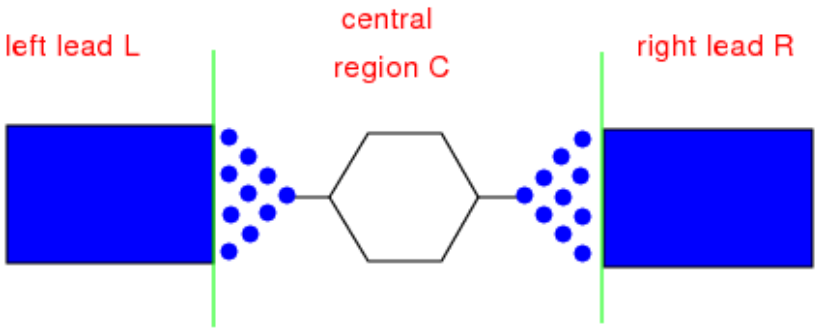
so far only few applications to simple systems

Example: 3-atom gold chain connected to two gold electrodes





Time-dependent transport: embedding technique



TD Kohn-Sham equation for orbitals

$$\left[i\partial_t - \begin{pmatrix} H_{LL}(t) & H_{LC} & 0 \\ H_{CL} & H_{CC}(t) & H_{CR} \\ 0 & H_{RC} & H_{RR}(t) \end{pmatrix} \right] \begin{pmatrix} \psi_{k,L}(t) \\ \psi_{k,C}(t) \\ \psi_{k,R}(t) \end{pmatrix} = 0$$

Time-dependent transport: embedding technique (cont.)

three equations

$$(i\partial_t - H_{LL}(t))\,\psi_{k,L}(t) = H_{LC}\psi_{k,C}(t) \qquad \text{(L)}$$

$$i\partial_t\psi_{k,C}(t) = H_{CL}\psi_{k,L}(t) + H_{CC}(t)\psi_{k,C}(t) + H_{CR}\psi_{k,R}(t) \qquad \text{(C)}$$

$$(i\partial_t - H_{RR}(t))\,\psi_{k,R}(t) = H_{RC}\psi_{k,C}(t) \qquad \text{(R)}$$

Retarded Green function for isolated lead $\alpha = L, R$

$$[i\partial_t - \hat{H}_{\alpha\alpha}(t)]g^R_\alpha(t,t') = \delta(t-t')$$

solve inhomogeneous Schrödinger equation (L) (simlarly for (R))

$$\psi_L = g^R_L\,[\text{r.h.s. of (L)}] + [\text{sol. of hom. SE } (i\partial_t - H_{LL}(t))\,\psi = 0]$$

$$\longrightarrow \psi_{k,L}(t) = \int_0^{t'} \mathrm{d}t'\; g^R_L(t,t')H_{LC}\psi_{k,C}(t') + ig^R_L(t,0)\psi_{k,L}(0)$$

Time-dependent transport: embedding technique (cont.)

Equation of motion for orbital projected on central region

$$[i\partial_t - \hat{H}_{CC}(t)]\psi_{k,C}(t) =$$

$$\int_0^t \mathrm{d}\bar{t}\, \Sigma^R_{emb}(t,\bar{t})\psi_{k,C}(\bar{t}) + \sum_\alpha H_{C\alpha} g^R_\alpha(t,0)\psi_{k,\alpha}(0)$$

with retarded embedding self energy

$$\Sigma^R_{emb}(t,t') = \sum_{\alpha=L,R} H_{C\alpha} g^R_\alpha(t,t') H_{\alpha C}$$

details in:

S. Kurth, G. Stefanucci, C.-O. Almbladh, A. Rubio, E.K.U. Gross, PRB **72**, 035308 (2005)

Time-dependent transport with TDDFT: some results

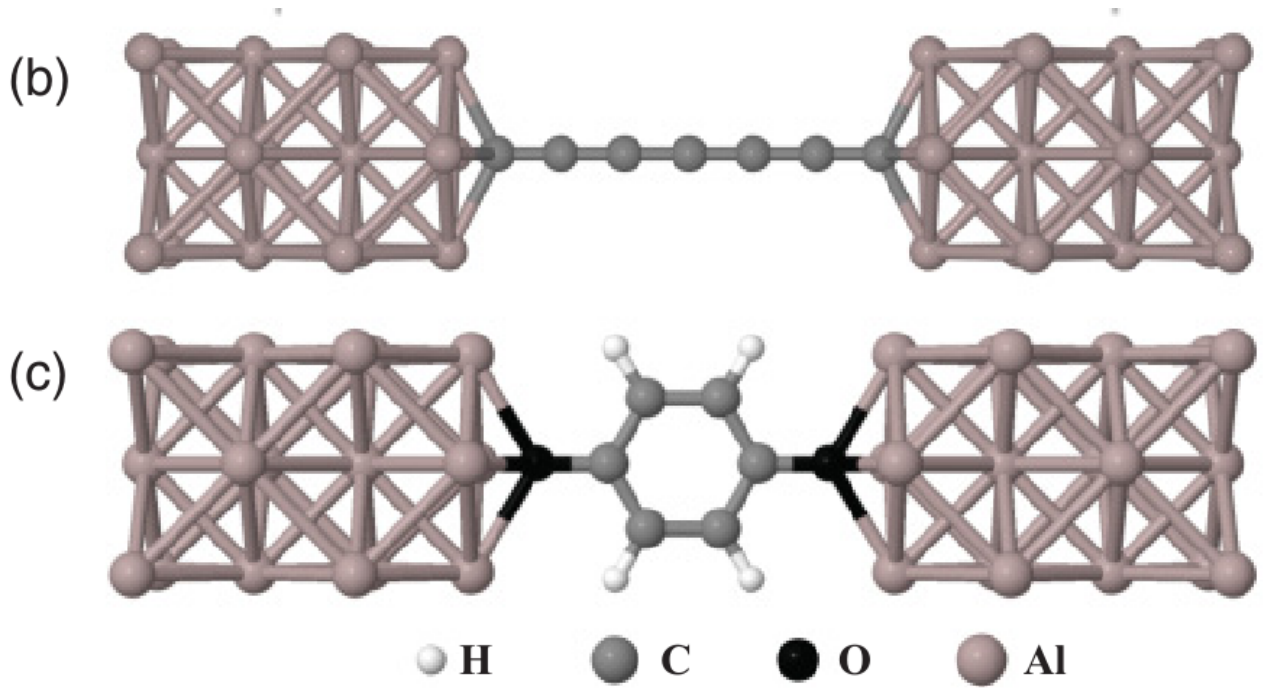
Time-dependent transport with adiabatic LDA

Reference:

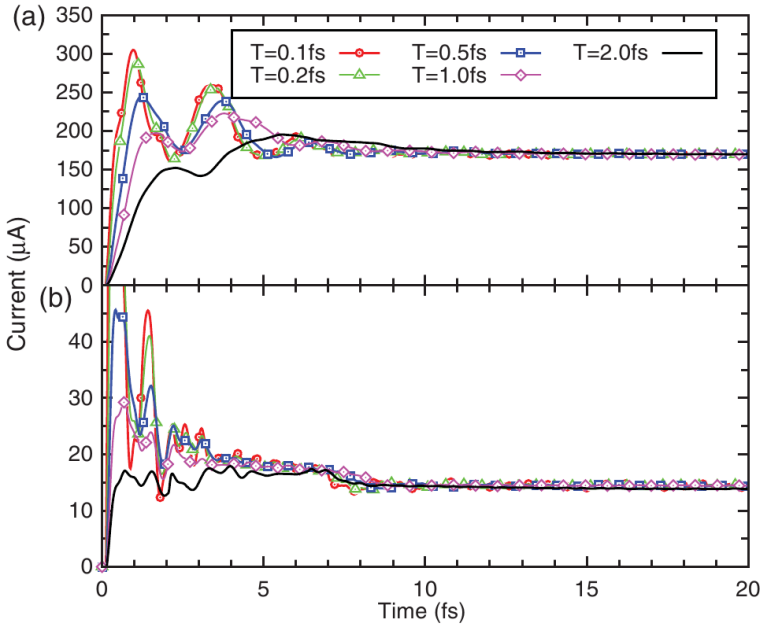
- C.-Y. Yam, X. Zheng, G.-H. Chen, Y. Wang, T. Frauenheim, T. Niehaus, PRB **83**, 245448 (2011)

Time-dependent transport with adiabatic LDA

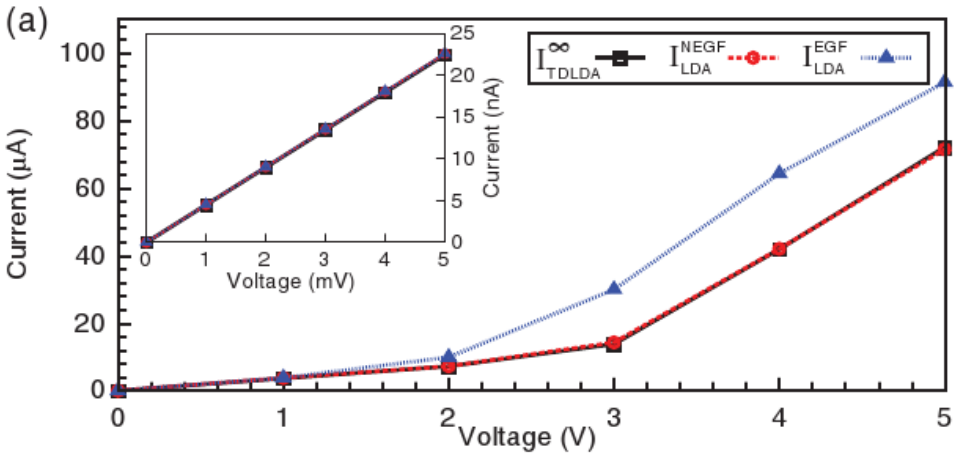
study of TD transport with variant of embedding scheme in adiabatic LDA



Time-dependent transport with adiabatic LDA

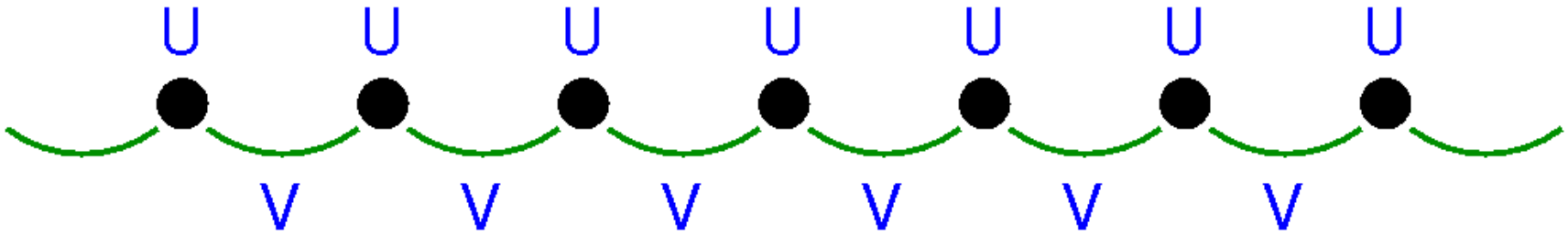


TD currents for different switching of the TD bias: $V(t) = V_0(1 - \exp(-t/T))$



steady-state currents extracted from TD calculations compared to results of Landauer formalism
$\longrightarrow$ currents agree

(Static) DFT for the Hubbard model



N.A. Lima et al (PRL **90**, 146402 (2003); EPL **60**, 601 (2002)):

parametrize total energy per site based on exact, Bethe ansatz (BA), solution of uniform Hubbard model with density n:

$$e^{BA}(n,U) = -\frac{2|V|\zeta}{\pi}\sin\left(\frac{\pi n}{\zeta}\right)$$

with parameter $\zeta(U)$ depending on interaction strength U
one can extract xc energy $e_{xc}^{BA}(n,U)$ from this parametrization

(Static) DFT for the Hubbard model

derivative discontinuity at $n = 1$

$$\Delta_{xc} = \lim_{\epsilon \to 0^+} [v_{xc}^{BALDA}(n = 1 + \epsilon) - v_{xc}^{BALDA}(n = 1 - \epsilon)]$$

$$= U - 4|V| \cos\left(\frac{\pi}{\zeta(U)}\right)$$

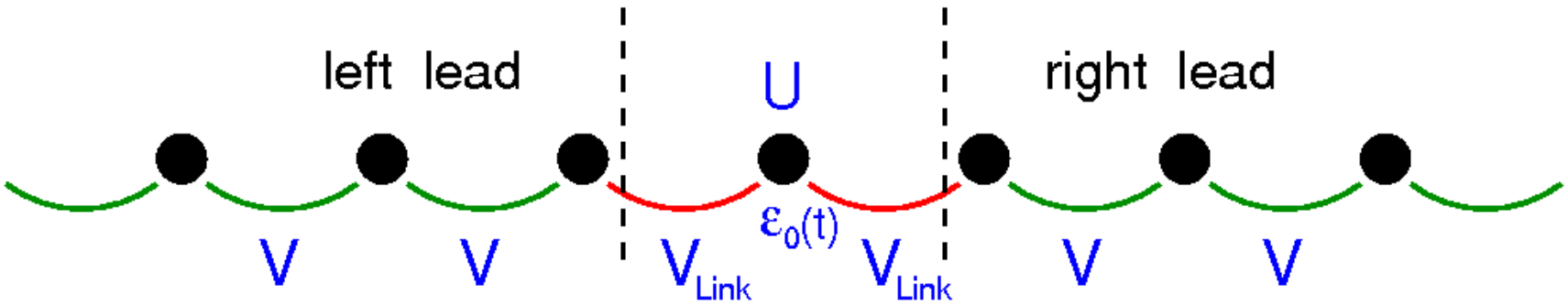
local approximation:

for non-uniform Hubbard models, i.e., non-constant on-site energies or even different interactions at each site:
use $e_{xc}^{BA}(n_i, U_i)$ as xc energy at site i (Bethe ansatz LDA, BALDA)

adiabatic approximation:

time-dependence of TDDFT xc potential at site i through $v_{xc}(i,t) = v_{xc}^{BALDA}(n_i(t))$ (C. Verdozzi, PRL **101**, 166401 (2008))

Simple impurity model for transport



one interacting impurity, Hubbard-like on-site interaction U, non-interacting leads, hopping V in leads and hopping V_{Link} from leads to impurity, (time-dependent) on-site energy $\varepsilon_0(t)$ at impurity

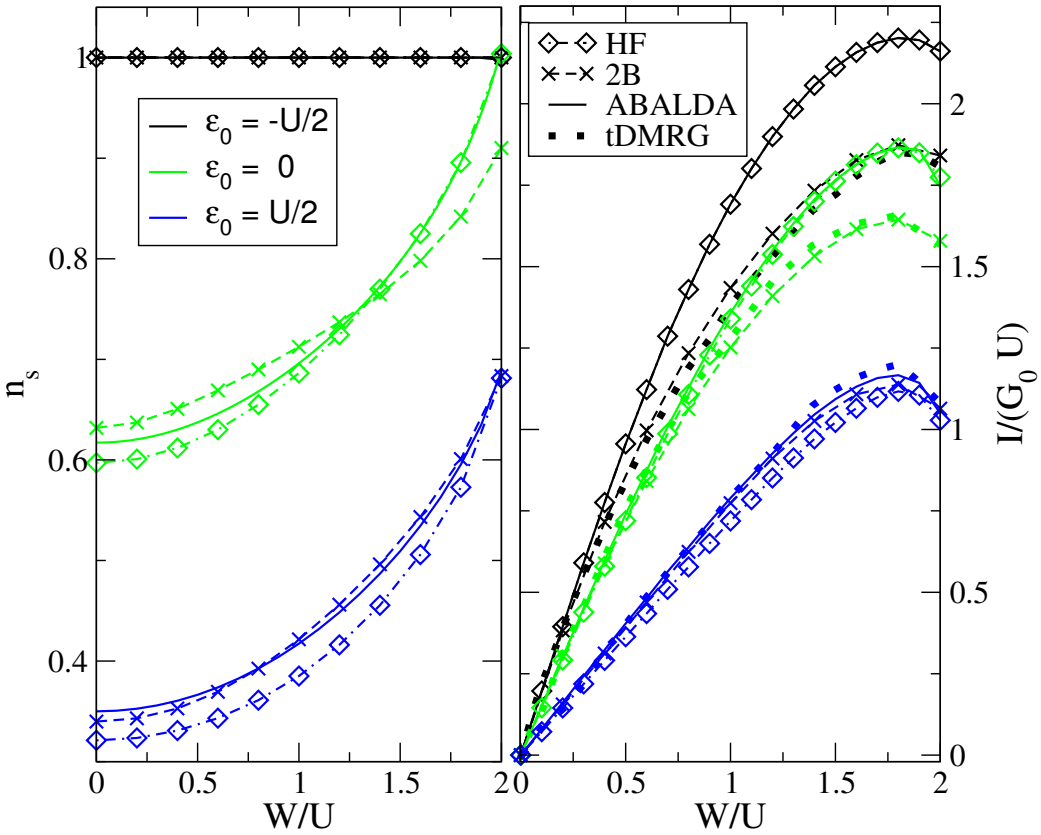
Comparison of TDDFT with many-body theory and DMRG

Reference:

- A.-M. Uimonen, E. Khosravi, A. Stan, G. Stefanucci, S. Kurth, R. van Leeuwen, E. K. U. Gross, PRB **84**, 115103 (2011)

Steady state densities and currents

Symmetric bias $W_L = -W_R = W/2$, $U = 1$, $V_{\mathrm{Link}} = 0.5$, $\varepsilon_F = 0$



HF: Hartree-Fock

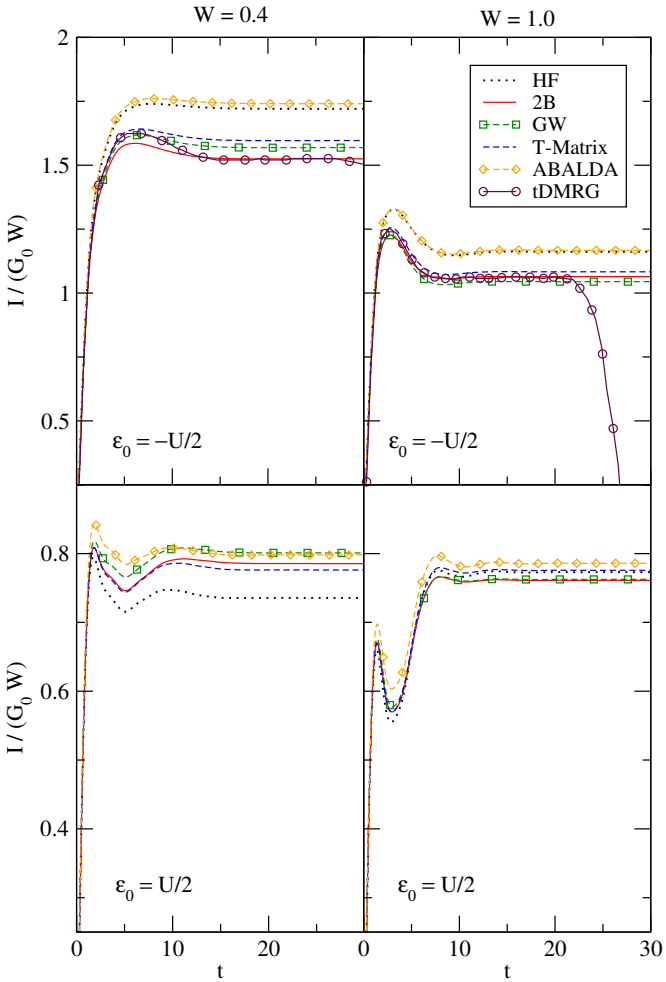
2B: second Born (Kadanoff-Baym eqs)

ABALDA: note: for local approximation, TDDFT corrections to Landauer vanish!

tDMRG: from Heidrich-Meisner et al PRB **79**, 235336 (2009)

for low bias, ABALDA currents reasonable, too large for high bias!

Time-dependent currents



$U = 0.5$, $V_{\mathrm{Link}} = 0.3535$

For higher onsite energy ε_0, ABALDA agrees better with many-body results

Dynamical Coulomb blockade and the derivative discontinuity

Reference:

- S. Kurth, G. Stefanucci, E. Khosravi, C. Verdozzi, E.K.U. Gross, PRL **104**, 236801 (2010); see also: C.A. Ullrich, Physics Viewpoint **3**, 47 (2010)

Self-consistency condition for steady state density

self-consistency condition for n

$$n = 2 \sum_{\alpha=L,R} \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} f_\beta(\omega - W_\alpha)\Gamma(\omega - W_\alpha)|G(\omega)|^2$$

$$G(\omega) = [\omega - v_{KS}(n) - \Sigma_L(\omega - W_L) - \Sigma_R(\omega - W_R)]^{-1}$$

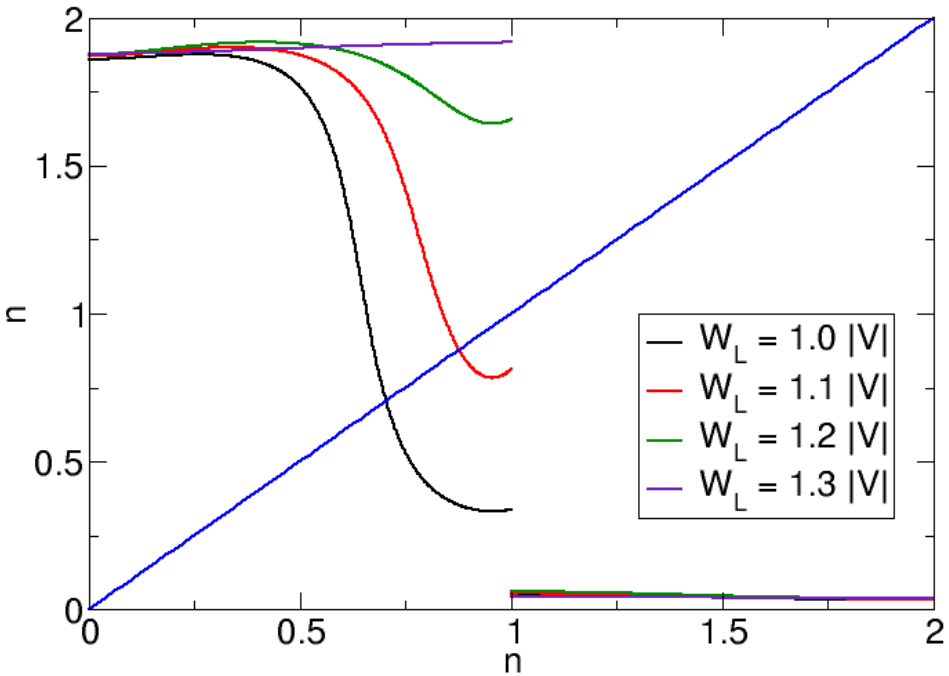
$$v_{KS}(n) = \varepsilon_0 + \frac{1}{2}Un + v_{xc}^{BALDA}(n)$$

Question:

What happens for an xc potential which is discontinuous?

Steady state self-consistent density for impurity model

l.h.s. and r.h.s. of self-consistency condition for n

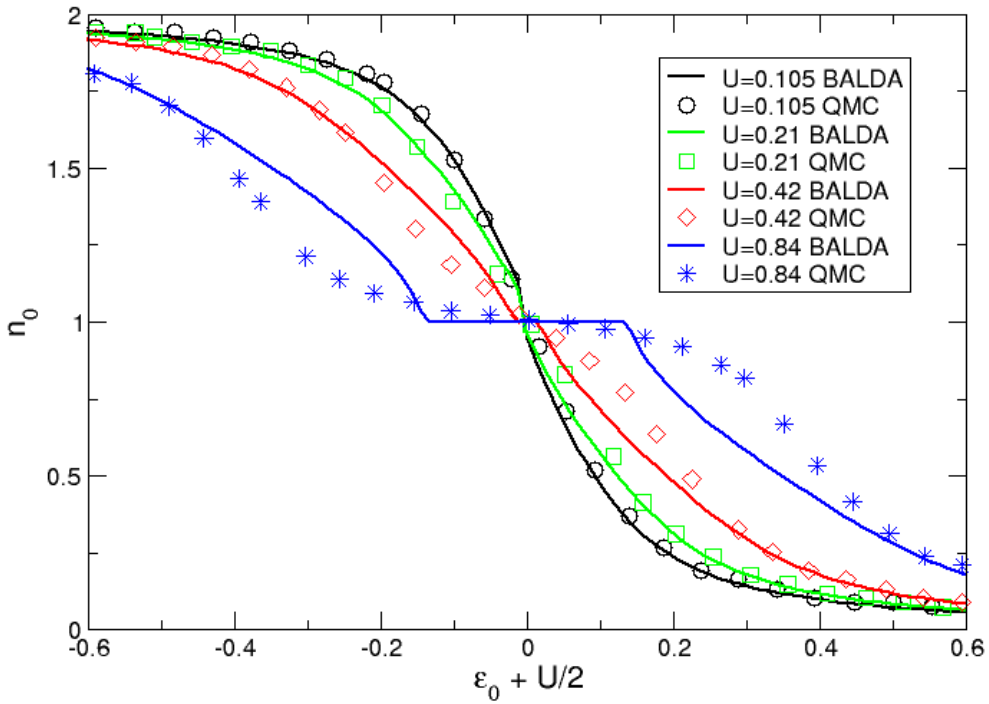


no solution for steady state density for some values of the bias.

to understand physics of this regime $\longrightarrow$ smoothen xc discontinuity

Ground state densities in BALDA and QMC

compare BALDA and QMC ground state densities of impurity model as function of the on-site energies ε_0 for different values of the interaction U; $V_{\mathrm{link}} = 0.18$

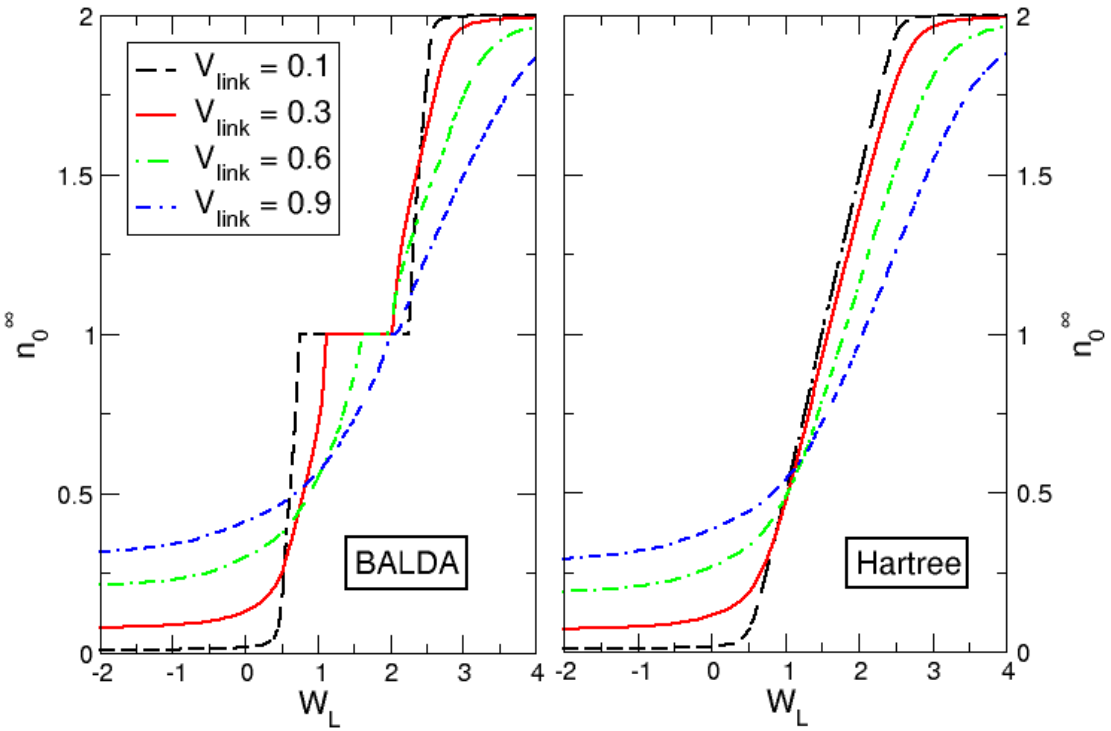


QMC results from:
X. Wang et al,
PRB **77**, 045119 (2008)

small U:
very good agreement
large U:
reasonable agreement + Coulomb blockade step

Steady-state density vs. bias

steady-state density as function of bias for different V_{Link}

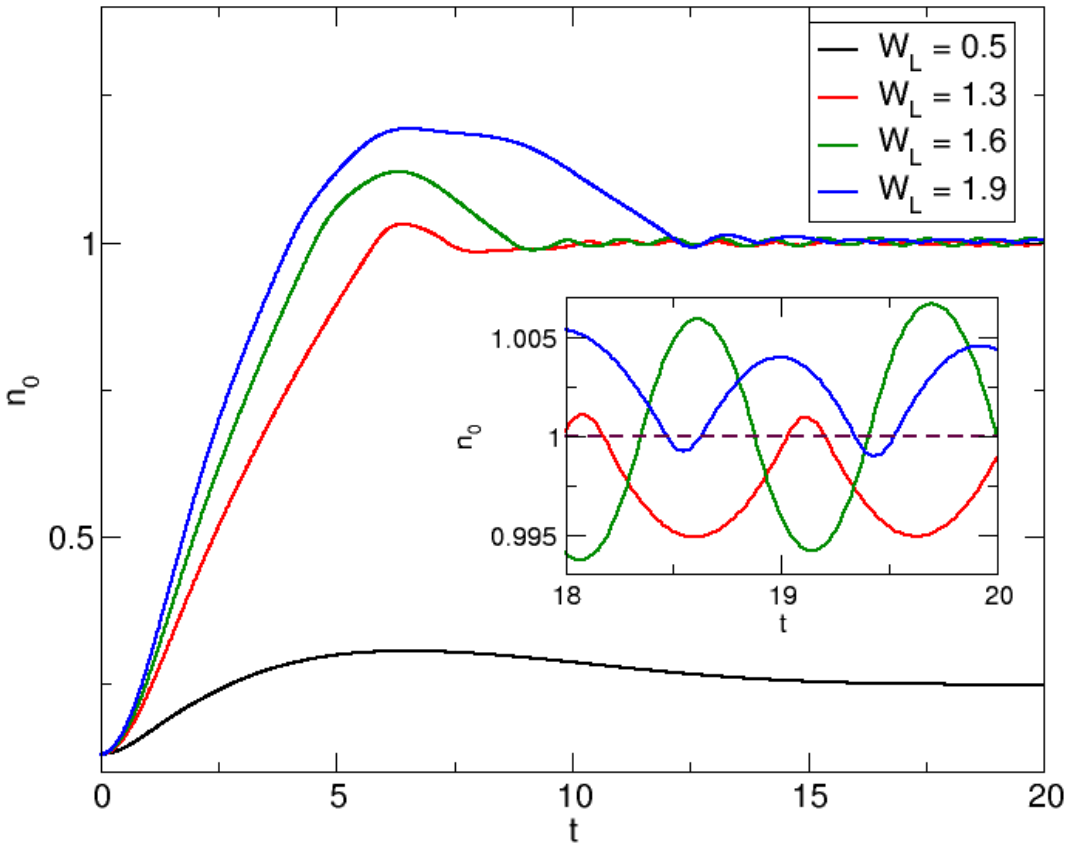


BALDA:
step structure
for small V_{link}
width of step: U
$\rightarrow$ Coulomb blockade
Hartree:
no step structure
$\rightarrow$ crucial role of
discontinuity

note: the role of the discontinuity in steady-state transport has also been discussed in C. Toher et al, PRL 95, 146402 (2005)

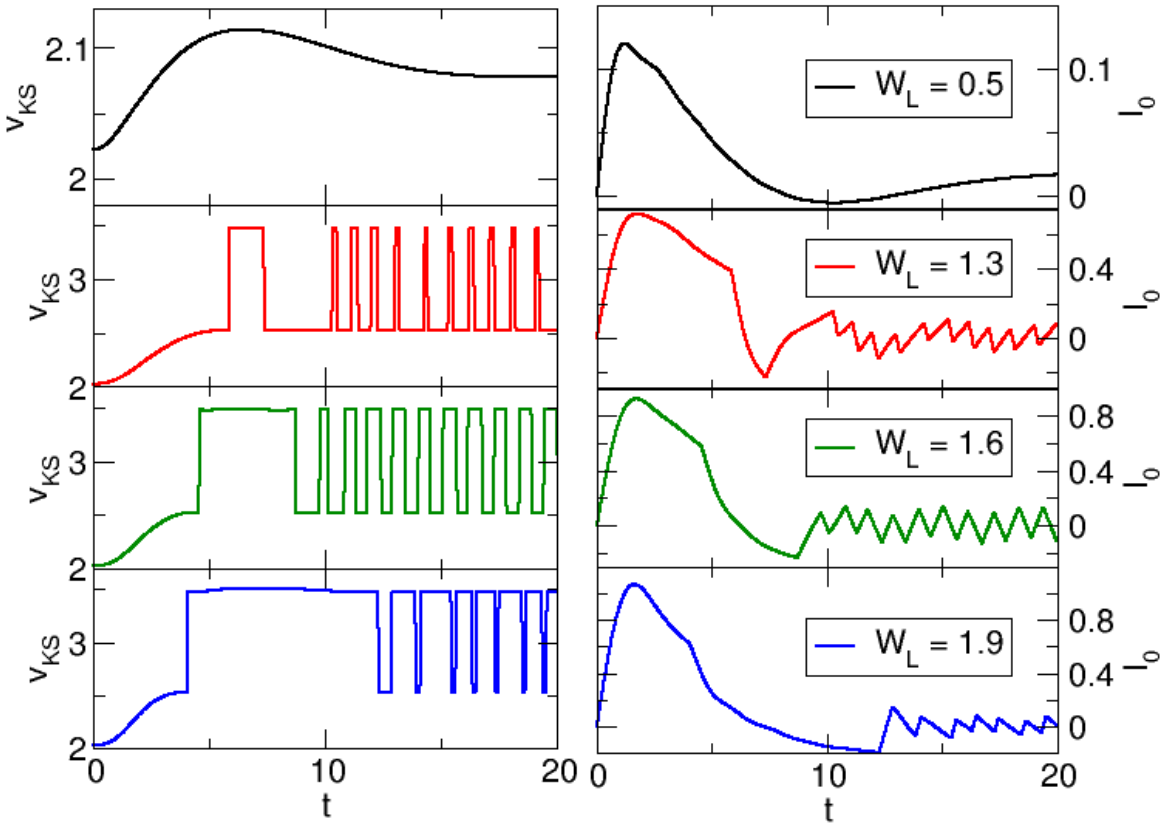
Time-dependent density in presence of discontinuity

Fermi energy $\varepsilon_F = 1.5|V|$, on-site energy $\varepsilon_0 = 2|V|$,
right bias $W_R = 0$, interaction $U = 2|V|$, $V_{\text{link}} = 0.3V$



for bias in step region of steady-state picture:
no steady state;
evolution towards a dynamic state of oscillating density around integer electron number

Time-dependent KS potentials and currents



in CB region:
KS potential rapidly varying; train of rectangular steps;
currents: sawtooth-like at impurity;

Bistabilities: switching between multiple steady states

References:

- E. Khosravi, A.-M. Uimonen, A. Stan, G. Stefanucci, S. Kurth, R. van Leeuwen, E. K. U. Gross, PRB **85**, 075103 (2012)
- A.-M. Uimonen, E. Khosravi, G. Stefanucci, S. Kurth, R. van Leeuwen, E.K.U. Gross, J. Phys.: Conf. Ser. **220**, 012018 (2010)

Self-consistency condition for steady state density

self-consistency condition for n

$$n = 2 \sum_{\alpha=L,R} \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} f_\beta(\omega - W_\alpha)\Gamma(\omega - W_\alpha)|G(\omega)|^2$$

$$G(\omega) = [\omega - v_{KS}(n) - \Sigma_L(\omega - W_L) - \Sigma_R(\omega - W_R)]^{-1}$$

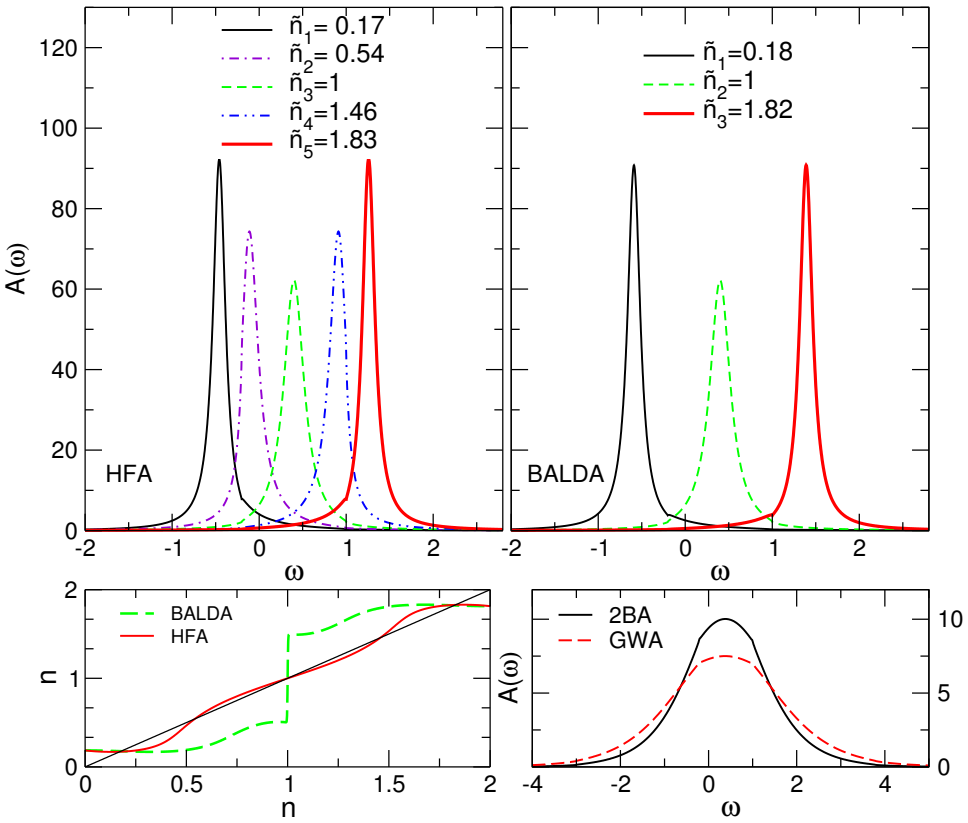
$$v_{KS}(n) = \varepsilon_0 + \frac{1}{2}Un + v_{xc}^{BALDA}(n)$$

note: this is a nonlinear equation which can have more than one solution, i.e., more than one steady state is possible!
Question: Can one switch between different steady states by applying an appropriate time-dependent external driving field?

Multiple steady state solutions: KS spectral functions

Parameters:

$U = 2,\ W_L = 1.8\ W_R = -1.0,\ V_{\mathrm{Link}} = 0.3,\ \varepsilon_0 = -0.6,\ \varepsilon_F = 0$

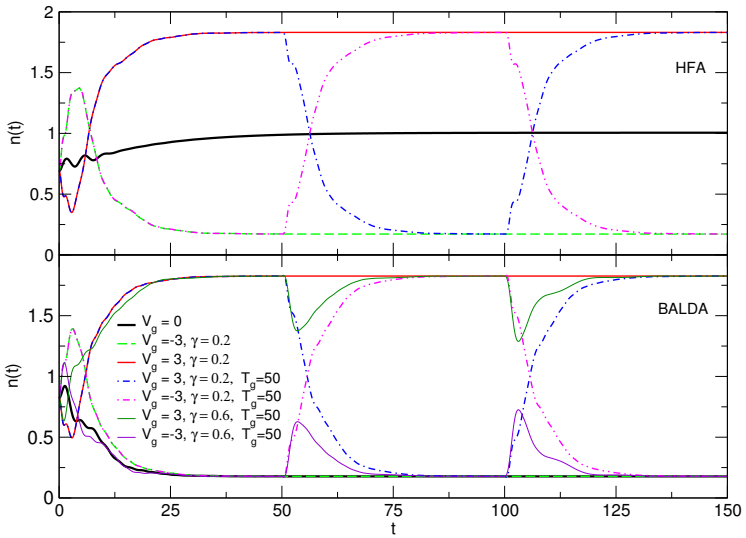


BALDA: 3 solutions
HF: 5 solutions
MBPT (GW,2B):
no closed equation for steady-state density;
no indications of multiple steady states beyond HF
note: MBPT gives much broader spectral functions (in steady state) than HF or BALDA

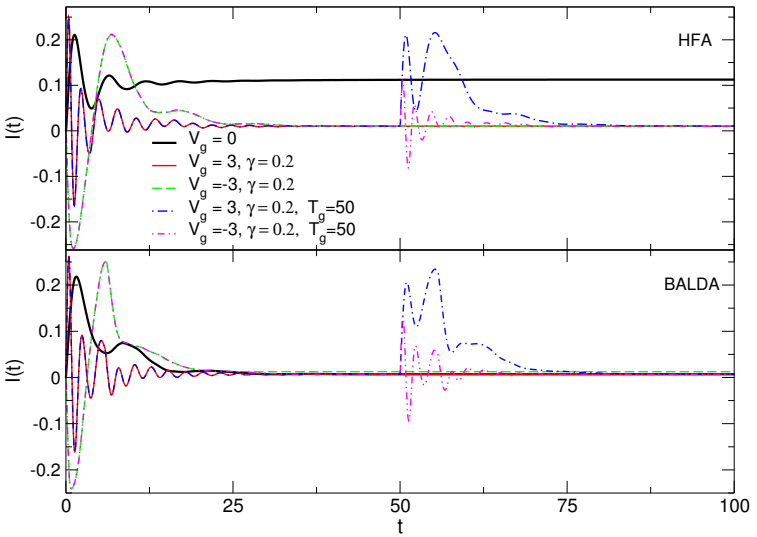
Time-dependent switching between different steady states

switching between steady states by time-dependent on-site energy

$$\varepsilon_0(t) = \begin{cases} V_g \exp(-\gamma t) & \text{if } 0 < t < T_g \\ -V_g \exp(-\gamma(t - T_g)) & \text{if } T_g < t < 2T_g \\ V_g \exp(-\gamma(t - 2T_g)) & \text{if } T_g < t < 2T_g \end{cases}$$



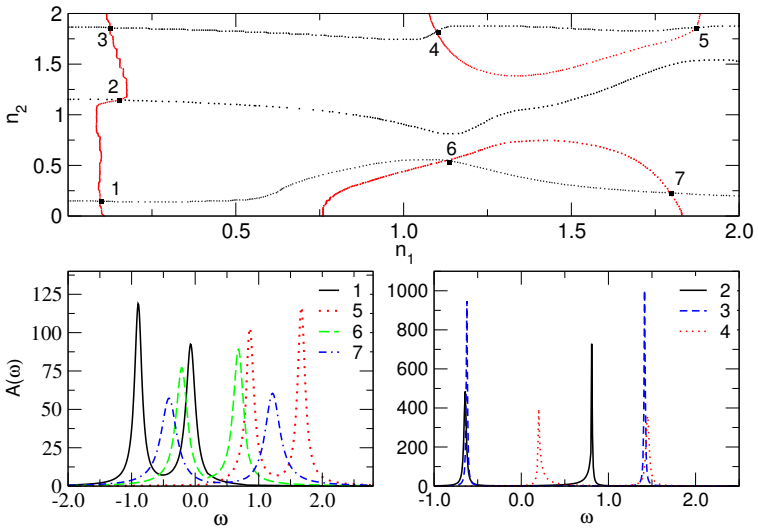
densities



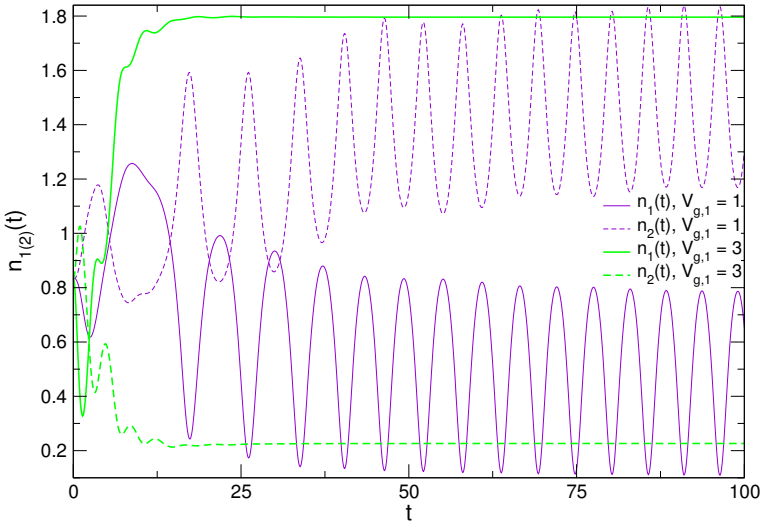
currents

Surprises with TD simulations

Two interacting Hubbard sites connected to leads in HF
bias and gate switched on at $t = 0$, use gate to drive system into different steady states; for certain gate parameters system can be driven into dynamical state with non-decaying oscillations



different steady states



time-evolution of densities

Summary and critique of adiabatic TDDFT for transport

- Various approaches to TDDFT for transport
  - Finite system approach
  - Master equation (open system)
  - Embedding scheme
- Some results of time-dependent transport with TDDFT
  - Adiabatic LDA TD transport calculations reach same steady state as given by Landauer+static DFT approach.
  - Adiabatic LDA can give reasonable TD and steady-state currents, especially in the low bias regime. MBPT at the level of second Born typically more accurate
  - Bistabilities and time-dependent switching between different steady states. But: No bistability found for correlated MBPT approaches. Is bistability an artefact of the adiabatic approximation?
  - Dynamical picture of Coulomb blockade. Again: Is this an artefact of the approximation?

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- Claudio Verdozzi, Univ. Lund