Quantum transport

Stefan Kurth

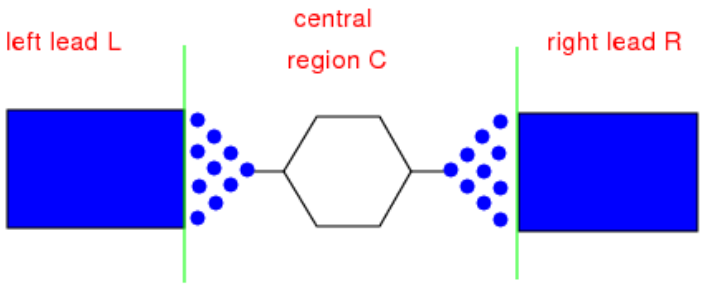
1. Universidad del País Vasco UPV/EHU, San Sebastián, Spain
2. IKERBASQUE, Basque Foundation for Science, Bilbao, Spain
3. European Theoretical Spectroscopy Facility (ETSF), www.etsf.eu

Outline

- Landauer steady-state approach to electron transport
- Critique of the Landauer approach
- TDDFT-based approaches to transport
  - Finite systems
  - Embedding scheme
  - Master equation
- Time-dependent transport phenomena
  - Bound-state oscillations
  - Bistability
  - Dynamical picture of Coulomb blockade
- Summary and critique of adiabatic TDDFT for transport

Landauer steady-state approach to electron transport

Standard approach: Landauer formalism plus static DFT



Starting point: Hamiltonian of static DFT in localized basis, define retarded Green function

$$\left[(E+i\eta)1-\begin{pmatrix} H_{LL} & H_{LC} & 0 \\ H_{CL} & H_{CC} & H_{CR} \\ 0 & H_{RC} & H_{RR} \end{pmatrix}\right]\begin{pmatrix} G_{LL} & G_{LC} & G_{LR} \\ G_{CL} & G_{CC} & G_{CR} \\ G_{RL} & G_{RC} & G_{RR} \end{pmatrix}=1$$

note: no direct hopping between left and right leads

Standard approach: Landauer formalism plus static DFT

project on central region

$$G_{CC}(E) = ((E + i\eta)1_{CC} - H_{CC} - \Sigma_L(E) - \Sigma_R(E))^{-1}$$

embedding self-energy for lead α

$$\Sigma_\alpha(E) = H_{C\alpha}\left((E + i\eta)1_\alpha - H_{\alpha\alpha}\right)^{-1} H_{\alpha C}$$

Transmission function

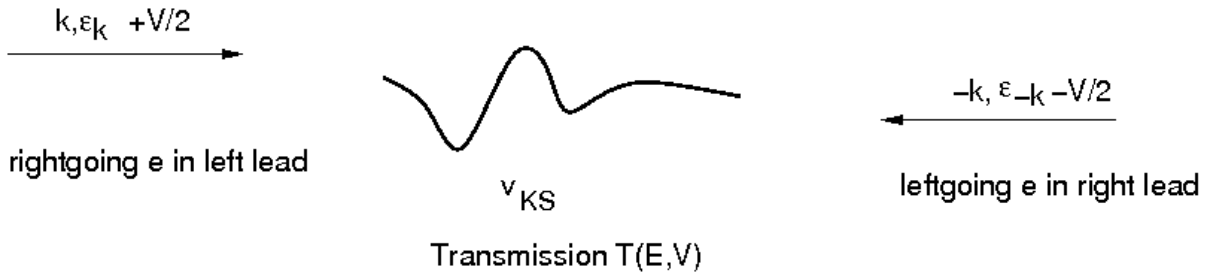
$$T(E,V) = \mathrm{Tr}\left\{\Gamma_L G_{CC} \Gamma_R G_{CC}\right\} \qquad \Gamma_\alpha = i(\Sigma_\alpha - \Sigma_\alpha^\dagger)$$

Landauer formula for steady-state current

$$I(V) = \frac{2e}{h}\int \mathrm{d}E\, T(E,V)\left(f_\beta(E + eV/2) - f_\beta(E - eV/2)\right)$$

Standard approach: Landauer formalism plus static DFT

Schematic idea of Landauer approach



Zero-bias conductance in Landauer formalism

$$\frac{G}{G_0} = \frac{dI}{dV}\bigg|_{V=0} = -\int \mathrm{d}E\, T(E) \frac{\partial f_\beta(E)}{\partial E} \qquad G_0 = \frac{2e^2}{h}$$

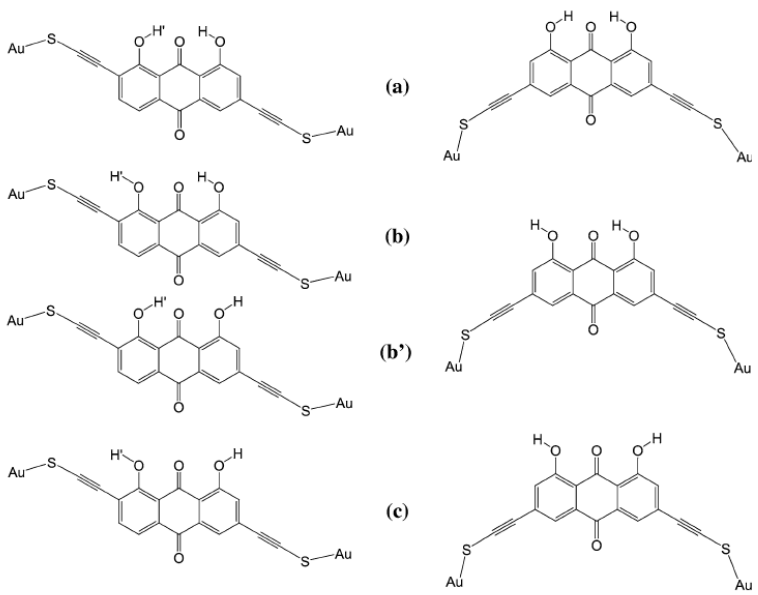
in zero-temperature limit: $\frac{G}{G_0} = T(E_F)$

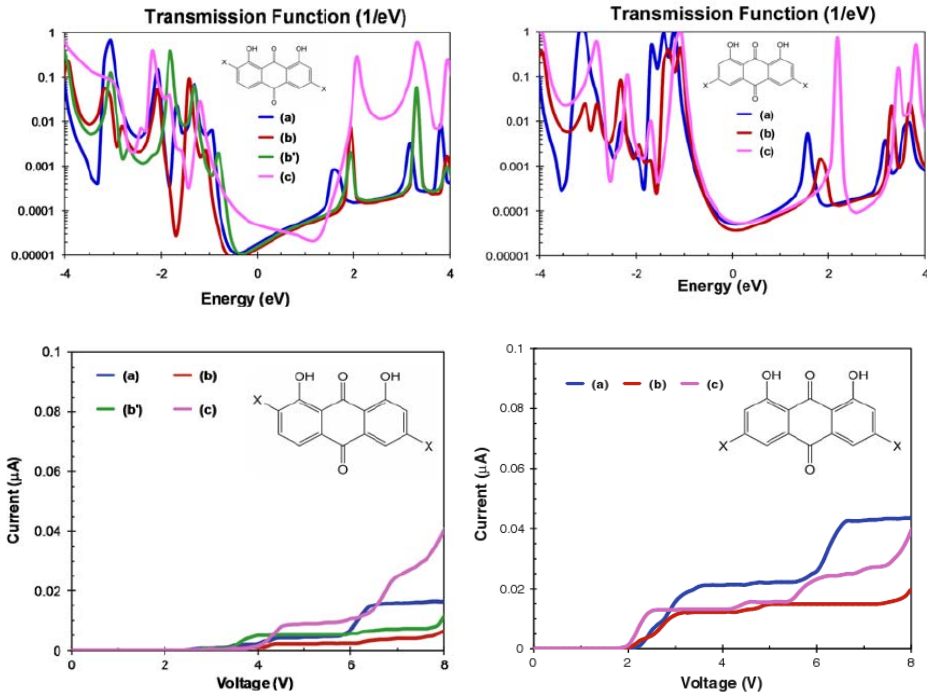
Example for application of Landauer formalism

Chrysazine molecule attached to gold leads

Ref.:

A. Zacarias, E.K.U. Gross, Theor. Chem. Acc. **125**, 535 (2010)





Critique of the Landauer approach

Empirical finding: currents through single (esp. organic) molecules often differ from experimental ones by 2-3 orders of magnitude

Basic assumptions behind Landauer:

- A stationary current always develops
- The stationary state is uniquely determined by the bias

Theoretical weaknesses:

- Landauer formalism vaild for non-interacting electrons
- Static DFT is a ground state theory and therefore in principle not suited to describe systems in a non-equilibrium situation
  $\longrightarrow$ even if exact effective KS potential of static DFT could be used, predictions of Landauer might still be incorrect!

$\longrightarrow$ use time-dependent DFT which is exact in principle.
Furthermore, it allows to study *time-dependent* transport phenomena (transients, AC bias, interaction with laser, etc...)

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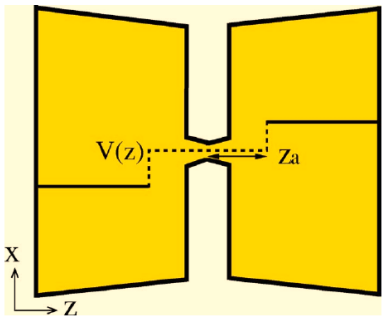
Approaches to time-dependent transport using TDDFT

Finite-system approach

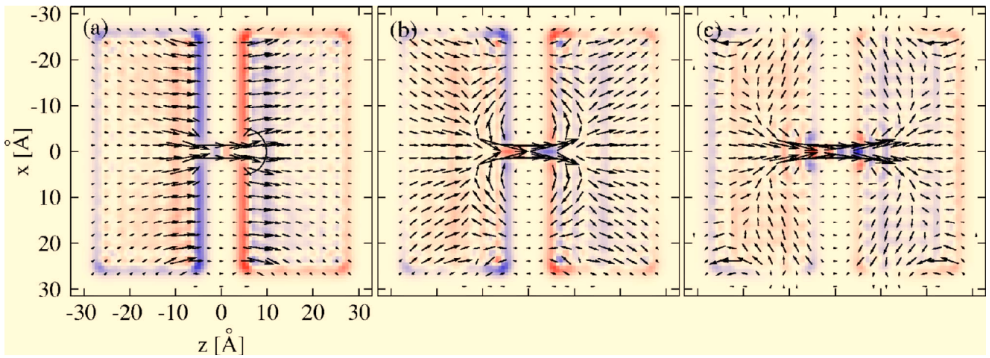
Idea: simulate nanosystem attached to large but finite leads



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

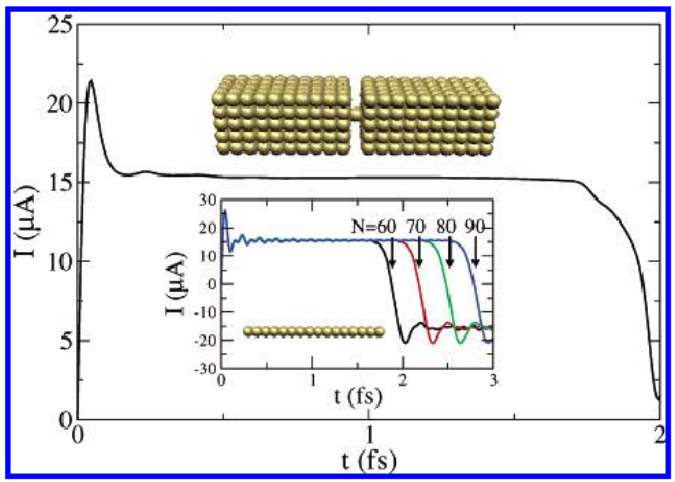
two large jellium leads connected by constriction (Sai et al, PRB **75**, 115410 (2007)): snapshots of current density in TDLDA



Finite-system approach (cont.)

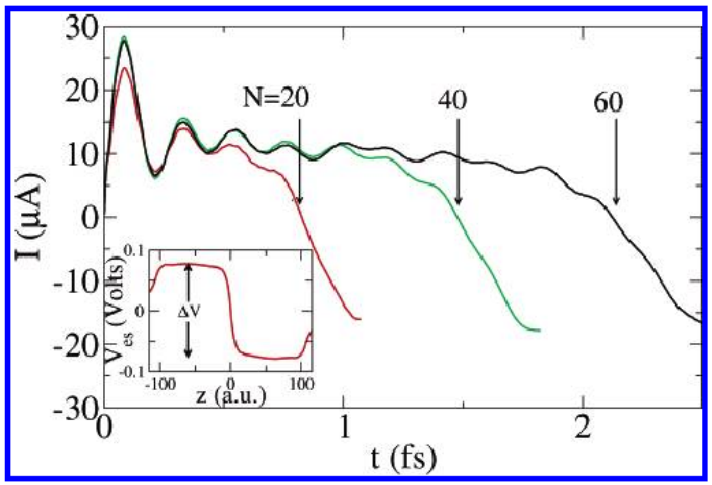
other example: tight-binding gold chain between gold electrodes (N. Bushong et al, Nano Lett. **5**, 2569 (2005))

non-interacting



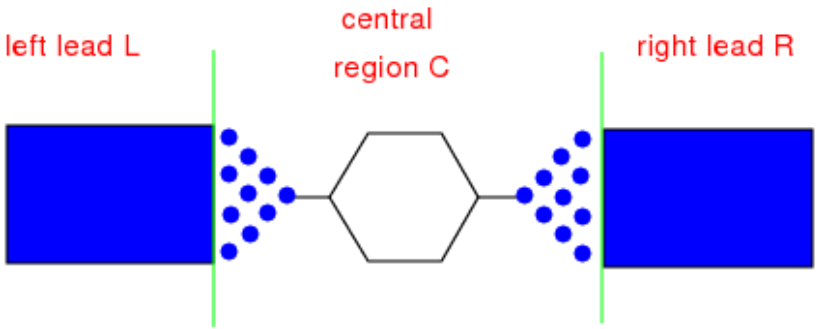
clear plateau in current after transients before eventually current dies out

TDLDA



current plateau still visible but somewhat less clear

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)

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 both $\longrightarrow$ 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

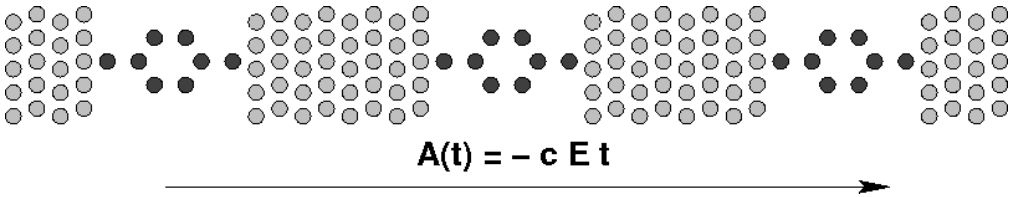
Master equation

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

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

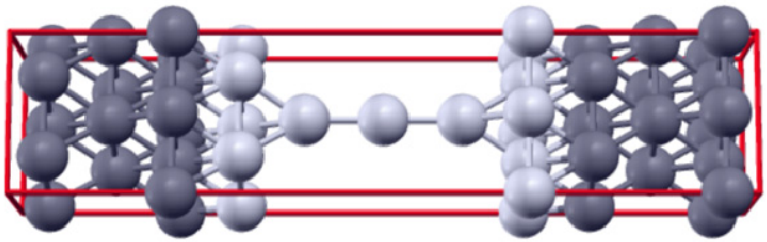
Quantum kinetic approach

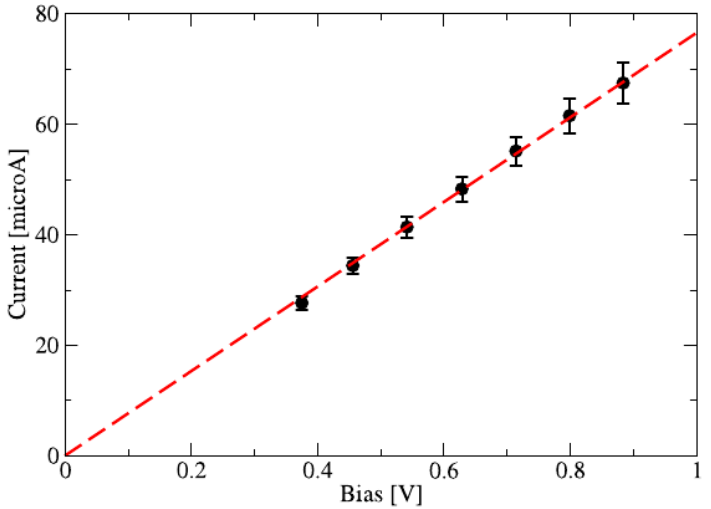
for practical convenience: use periodic boundary conditions, i.e., schematically as



so far only few applications to simple systems

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





Time-dependent transport phenomena

Bound-state oscillations for non-interacting electrons

G. Stefanucci (PRB **75**, 195115 (2007)): If Hamiltonian of a (noninteracting) biased system in the long-time limit supports two or more bound states $\longrightarrow$ total current in long-time limit

$$\lim_{t\to\infty} I_\alpha(t) = I_\alpha^{(S)} + I_\alpha^{(D)}(t)$$

with steady-state part $I_\alpha^{(S)}$ and dynamical part

$$I_\alpha^{(D)}(t) = 2\sum_{b,b'} f_{b,b'}\Lambda^\alpha_{b,b'}\sin((\varepsilon_b - \varepsilon_{b'})t)$$

where sum runs over the bound states of the Hamiltonian in long-time limit

Remarks: 1) $f_{b,b'}$ depends on history of TD Hamiltonian
2) No steady state in long-time limit (violation of one of the assumptions underlying Landauer)

Bound-state oscillations in density

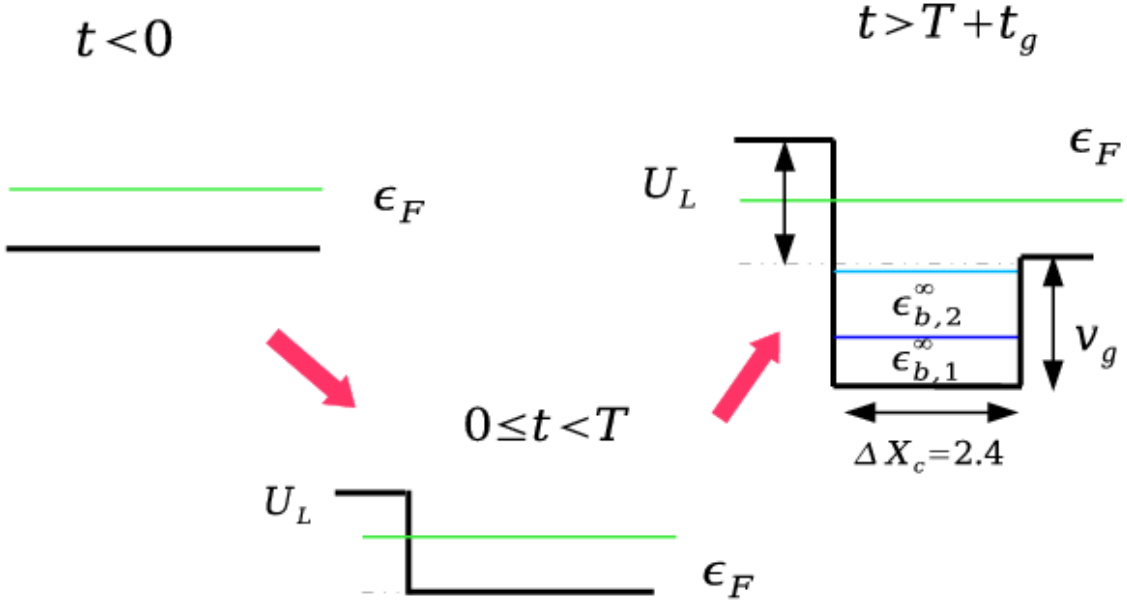
current and density related via continuity equation
$\longrightarrow$ also dynamical contribution to density for large t

$$n^{(D)}(\mathbf{r},t) = \sum_{b,b'} f_{b,b'} \cos[(\varepsilon_b^\infty - \varepsilon_{b'}^\infty)t]\psi_b^{\infty *}(\mathbf{r})\psi_{b'}^\infty(\mathbf{r})$$

note:
the terms $b = b'$ lead to a history-dependent contribution (through $f_{b,b}$) to the time-independent part of the density

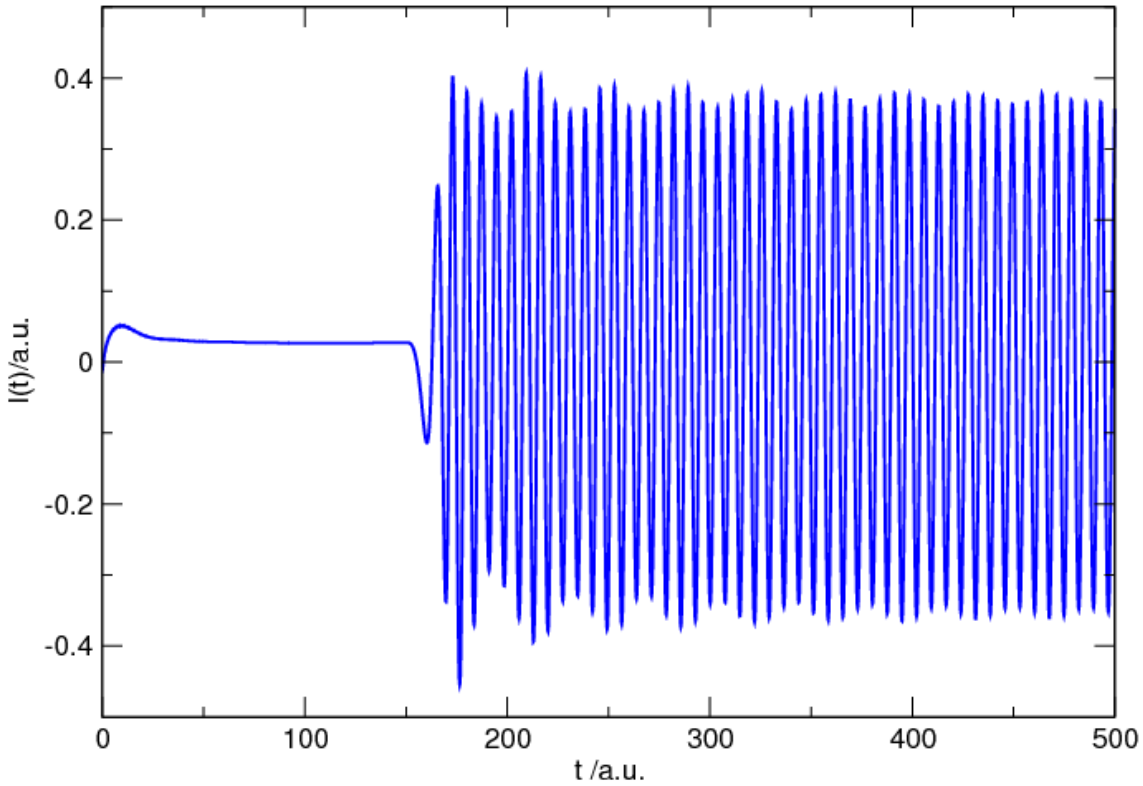
Bound-State Oscillations in One-Dimensional Model

Start with constant potential, at $t = 0$ apply sudden DC bias in left lead, wait until steady state develops, then switch on gate potential (with switching time t_g) in device region which creates two bound states



Refs.: E. Khosravi et al, Appl. Phys. A **93**, 355 (2008); Phys. Chem. Chem. Phys. **11** 4535 (2009)

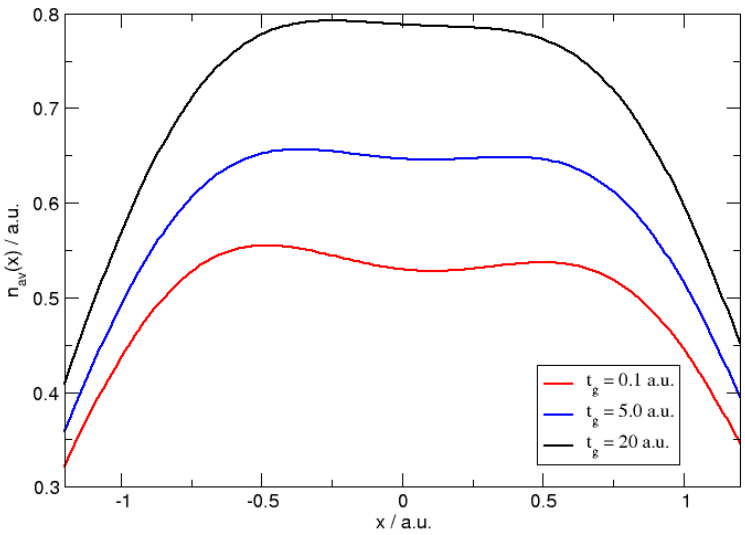
1D model: time-dependent current



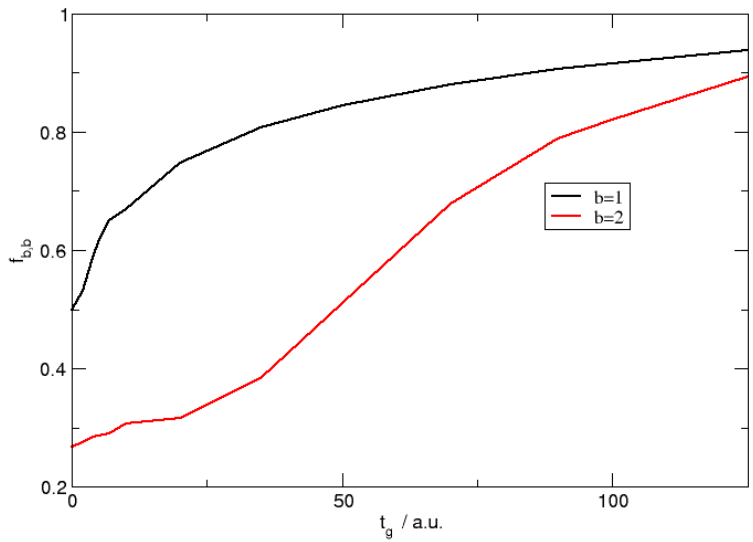
note:

amplitude of current oscillations large compared to steady-state current

1-D Model: Bound-State Density and Occupation



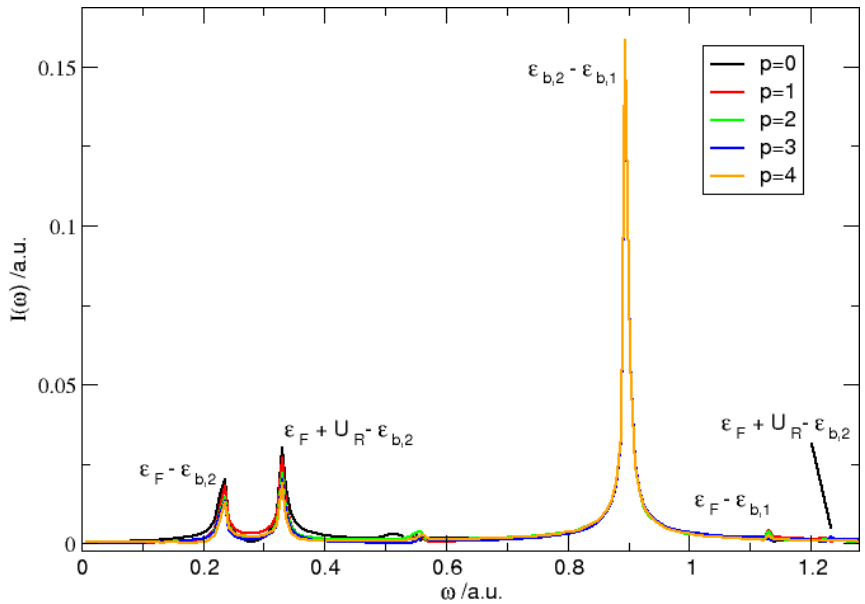
bound-state contribution to time-independent density for different switching times



occupation numbers $f_{b,b}$ of bound states as function of switching time

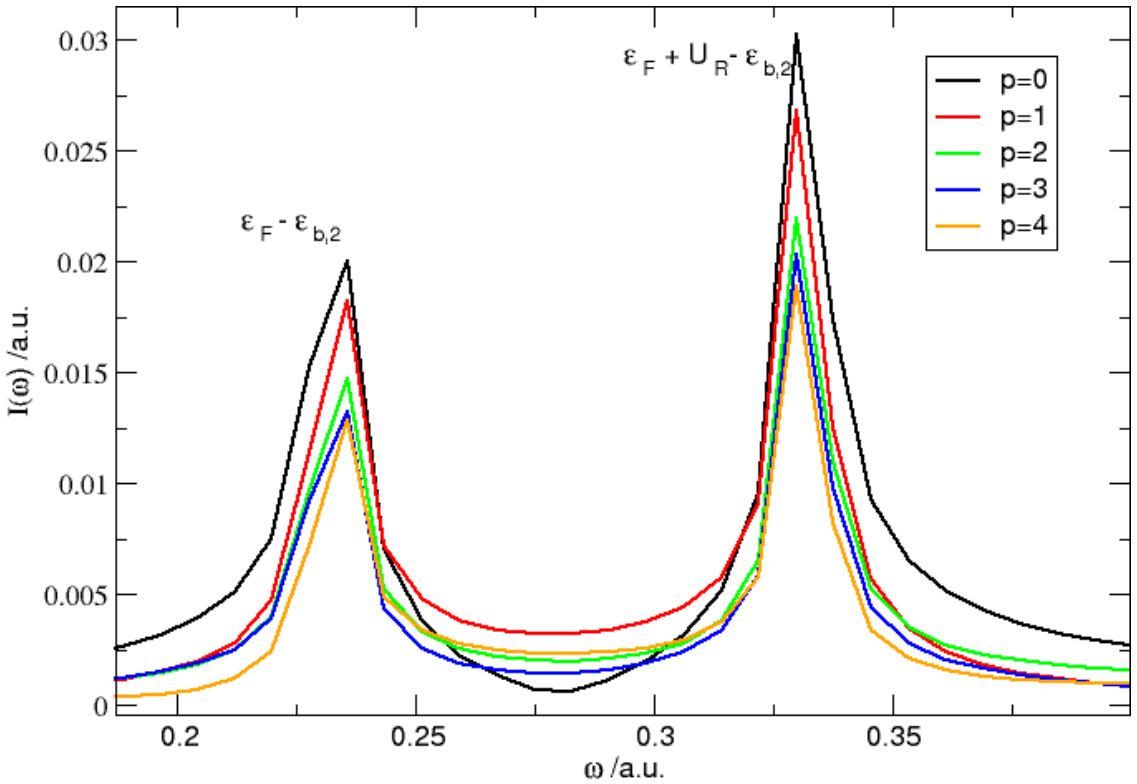
1D model: transients in current

biased system with two bound states in initial state: Fourier transform of current for finite time interval of length $T_0 = 800$ a.u. but different starting points $t_p = (2 + p) \times 100$ a.u.



note: not only transition between bound states but also between bound states and continuum

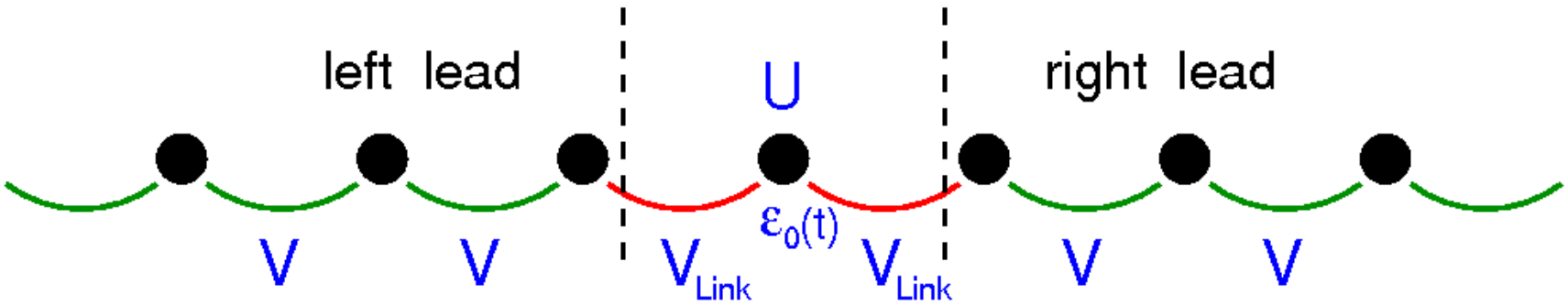
1D model: transients in current



note:

bound-continuum transitions die out slowly ($\sim 1/t$)

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

Simple impurity model: steady-state condition in DFT

use local KS potential, apply DC bias W_α in lead $\alpha = L, R$, assume there exists steady state with density n at impurity
$\longrightarrow$ self-consistency condition for n

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

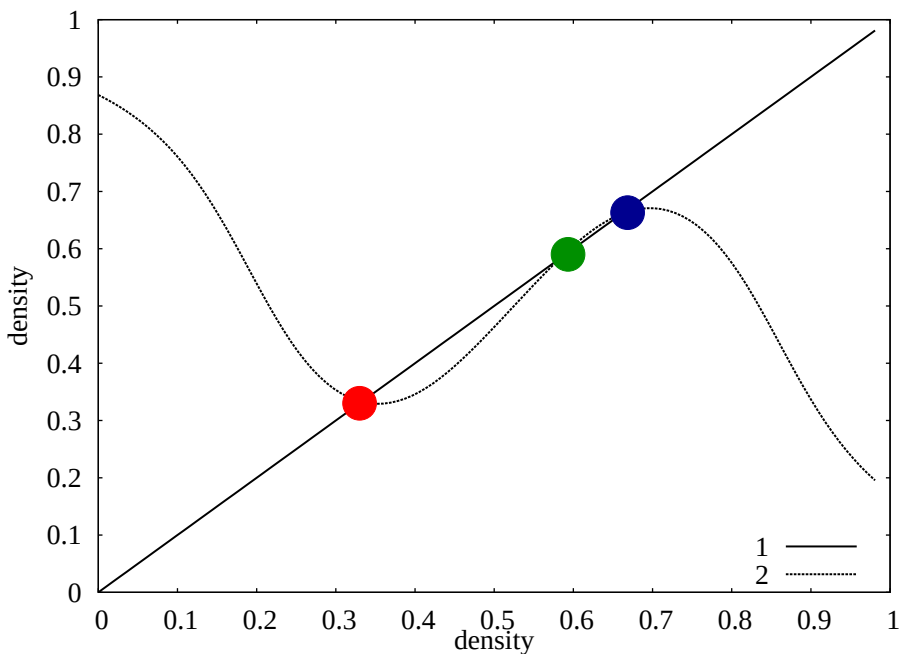
$$G(\omega) = [\omega - v_{KS}(n) - \Sigma(\omega - W_L) - \Sigma(\omega - W_R)]^{-1}$$

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

$f_\beta(\omega)$: Fermi function at inverse temperature β
$\Sigma_\alpha(\omega)$: embedding self energy for lead α

Multiple solutions for steady-state condition in DFT

lead bands from $-2|V|$ to $2|V|$, Fermi energy $\varepsilon_F = 0.6|V|$, on-site interaction $U = 4|V|$, hopping to impurity $V_{\mathrm{Link}} = 0.7V$, on-site energy $\varepsilon_0 = 0$, right bias $W_R = 0$, left bias $W_L = 3|V|$
use Hartree approximation (which for our model is equivalent to Hartree-Fock): $v_{xc} \equiv 0$

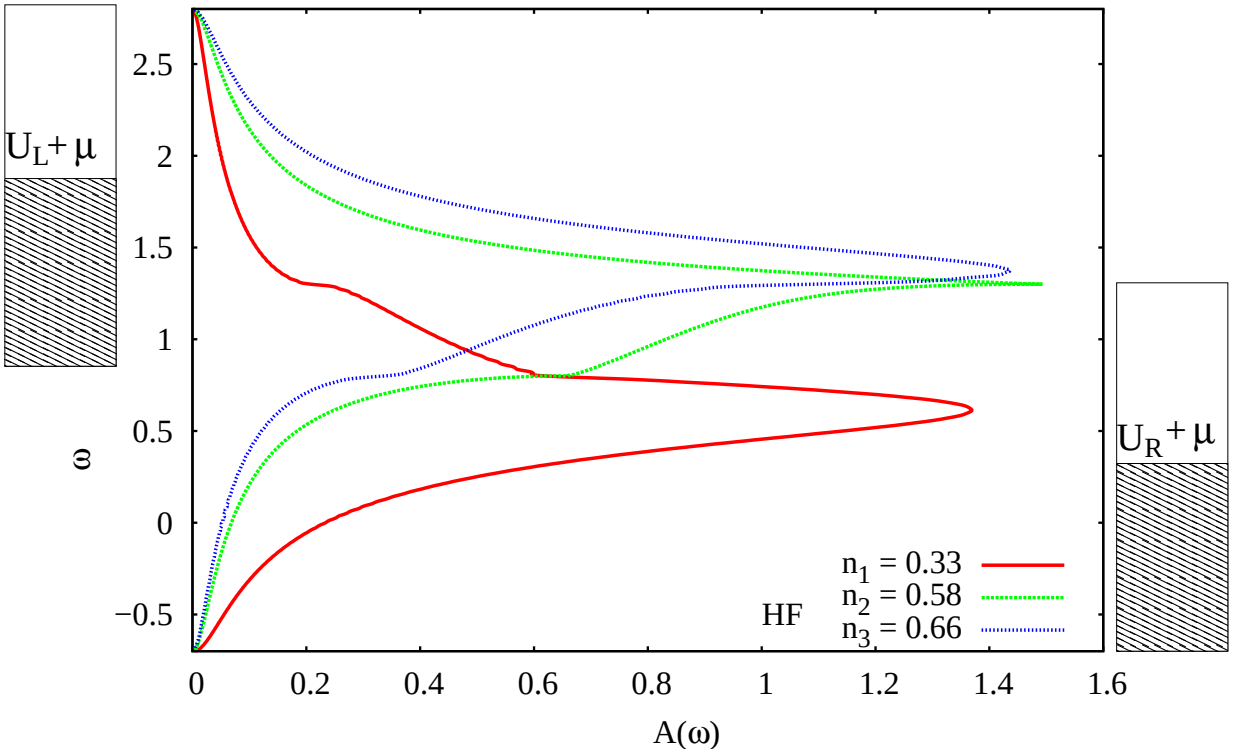


three solutions for self-consistent density

$$n_1 = 0.33$$
$$n_2 = 0.58$$
$$n_3 = 0.66$$

Question: which steady state can be reached by time propagation?

Spectral functions for multiple steady-state solutions

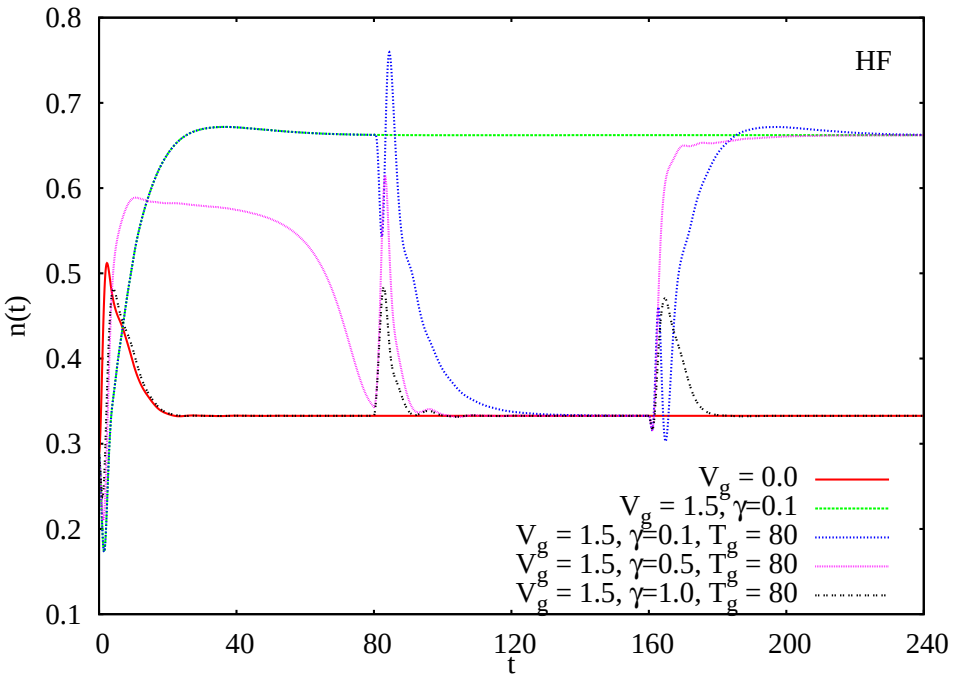


spectral function for n_1 peaked in energy range of right band, for n_3 peaked in range of left band, for n_2 peaked at top of right band

Time-dependent switching between different steady states

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

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

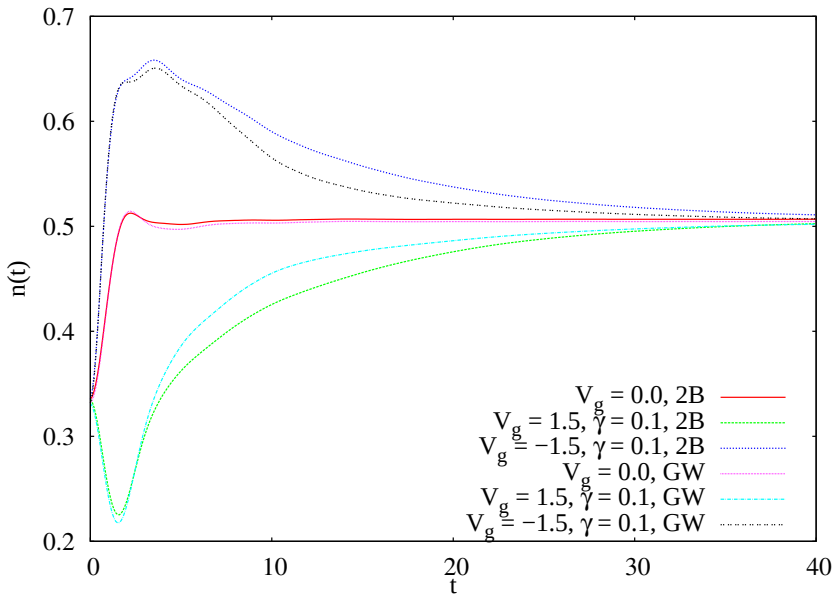


second steady state (density n_2) unstable, cannot be reached by time-propagation

Inclusion of correlation in GW and second Born

BUT: using diagrammatic techniques (Kadanoff-Baym equations) we found that inclusion of correlation beyond HF such as GW or second Born approximation tends to destroy mustistability

TD density in GW and 2B



Refs.:

- A.-M. Uimonen et al, J.Phys: Conf. Ser. **220**, 012018 (2010)
- E. Khosravi et al, arXiv:cond-mat/1112.2871 (2011)

Local functional with derivative discontinuity

use a modified version of the functional suggested by Lima et al, PRL **90**, 146402 (2003) based on the Bethe ansatz for the uniform Hubbard model (Bethe-ansatz LDA, BALDA)

important property: derivative discontinuity at half filling

derivative discontinuity at $n = 1$

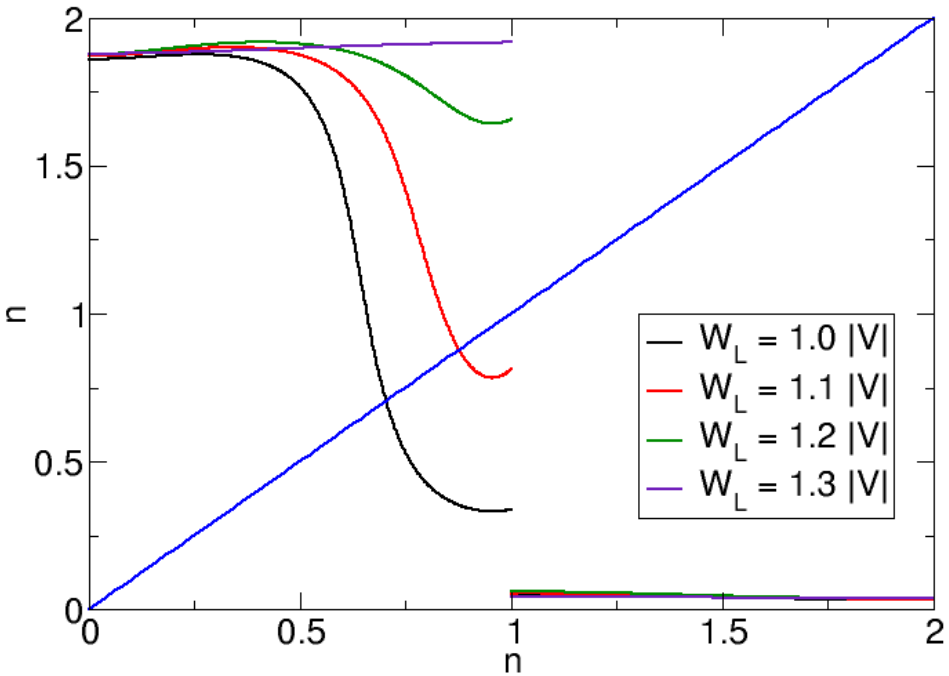
$$\Delta = \lim_{\epsilon \to 0^+} [v_{xc}^{BALDA}(n = 1 + \epsilon) - v_{xc}^{BALDA}(n = 1 - \epsilon)]$$

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

where $\zeta(U)$ parametrically depends on the on-site interaction U

Steady state self-consistent density for impurity model

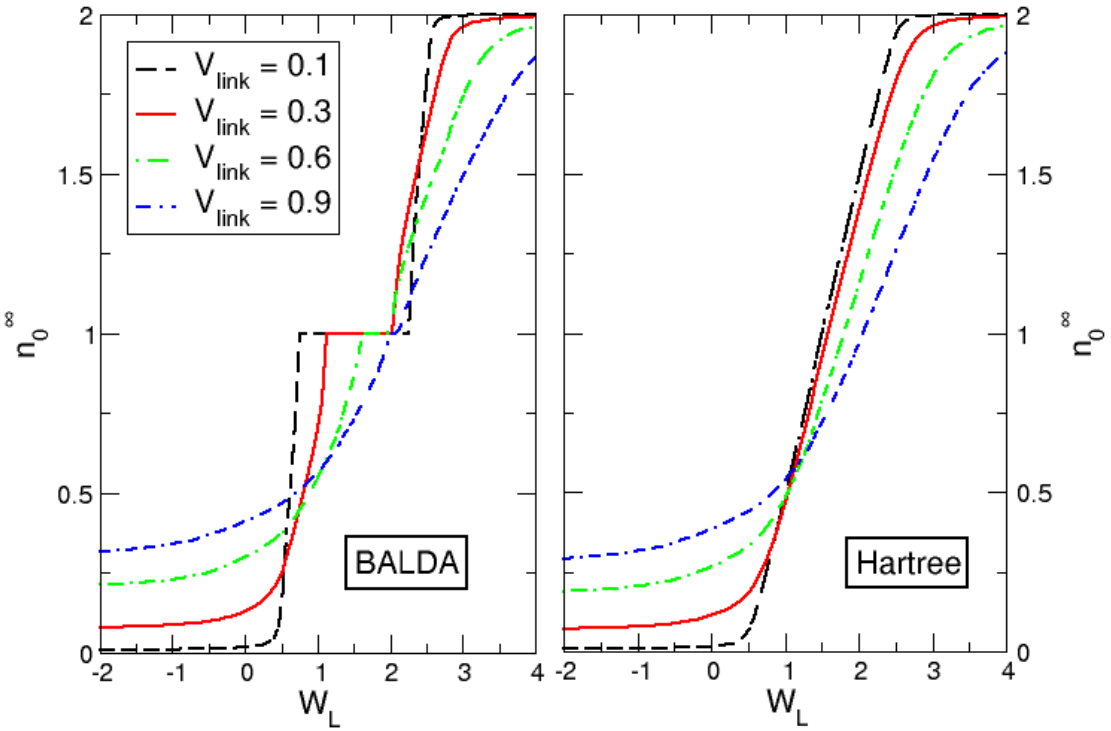
l.h.s. and r.h.s. of self-consistency condition for n



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

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

Steady-state density vs. bias for different V_{Link}



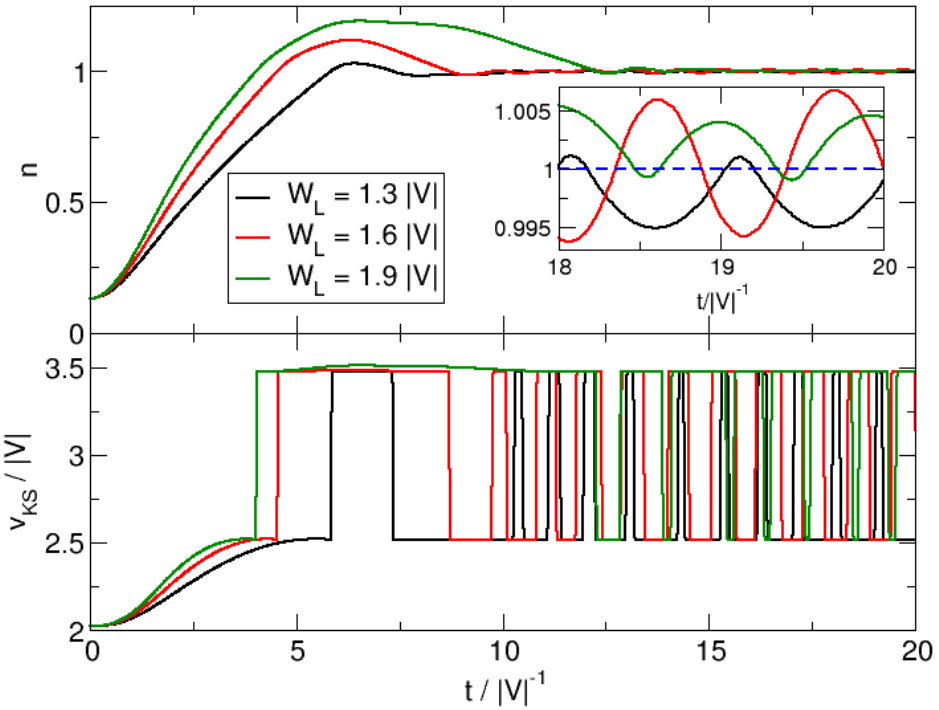
BALDA:
step structure
for small V_{Link}
width of step: U
$\longrightarrow$ Coulomb blockade

Hartree:
no step structure
$\longrightarrow$ 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)

Dynamical picture of Coulomb blockade

TD density and KS potential in presence of discontinuity



density shows small oscillations around integer occupation

TD KS potential: series of almost rectangular potential steps

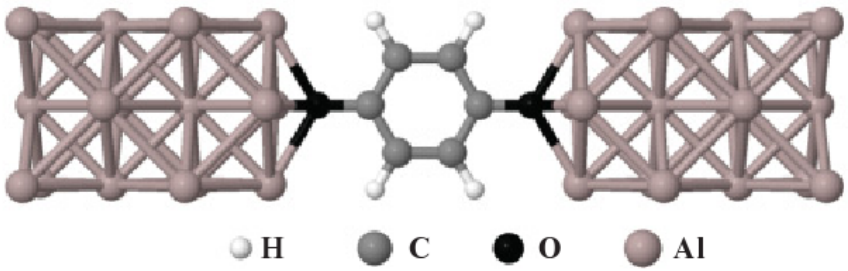
Ref: S. Kurth, G. Stefanucci, E. Khosravi, C. Verdozzi, E.K.U. Gross, PRL **104**, 236801 (2010);
see also: C.A. Ullrich, Physics **3**, 47 (2010)

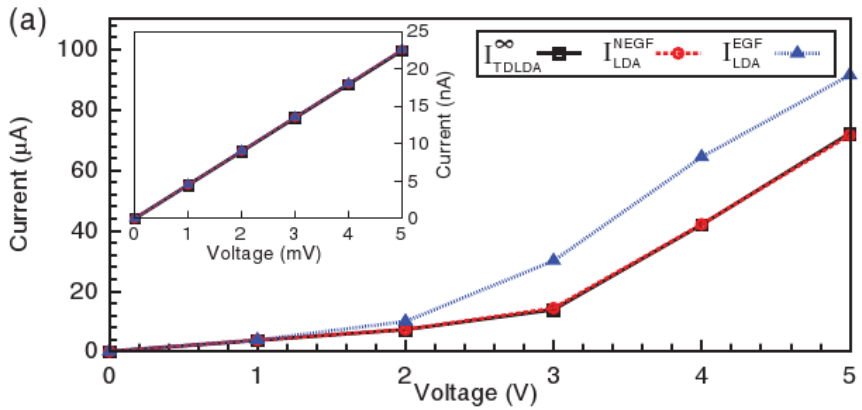
Summary and critique of adiabatic TDDFT for transport

- Standard Landauer + static DFT approach in principle incomplete
- TDDFT approach to transport in principle ok
- Various approaches to TDDFT for transport
  - Finite system approach
  - Embedding scheme
  - Master equation (open system)
- Time-dependent phenomena in transport
  - Bound-state oscillations for non-interacting electrons (no steady state)
  - Bistabilities and time-dependent switching between different steady states
  - Dynamical picture of Coulomb blockade

Summary and critique of adiabatic TDDFT for transport

- TDLDA does not give any corrections to the Landauer formula





from C.-Y. Yam et al, PRB **83**, 245448 (2011)

- Bistability has only been found for adiabatic functionals, i.e., functionals without memory but not for correlated MBPT approaches beyond HF. Is bistability an artefact of the adiabatic approximation?