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Dynamical Coulomb blockade and the derivative discontinuity: a not-so-steady state

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- C. Verdozzi, Univ. Lund, Sweden

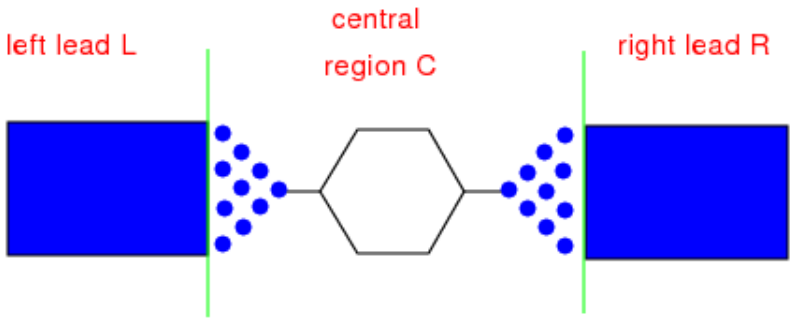
Outline

- Time-dependent density functional theory for transport
- Derivative discontinuity in static DFT
- A simple impurity model
- Transport through impurity model: time-dependent picture of Coulomb blockade
- Summary

Time-Dependent Description of Transport: Why and How?

- transport is an inherent non-equilibrium phenomenon
- steady state typically achieved at the end of an evolution process
- can describe TD phenomena: transients, TD bias, external TD fields, ...
- method: time-dependent DFT: in principle exact

TDDFT for transport



TD Kohn-Sham equation for orbitals

$$[i\partial_t - \hat{H}(t)]\psi_k(t) = 0$$

Hamiltonian of extended system L-C-R, no direct hopping between left and right leads

$$\hat{H}(t) = \begin{pmatrix} H_{LL}(t) & H_{LC} & 0 \\ H_{CL} & H_{CC}(t) & H_{CR} \\ 0 & H_{RC} & H_{RR}(t) \end{pmatrix}$$

Time-Dependent Density Functional Theory

density from orbitals

$$n(\mathbf{r},t) = \sum_{k}^{occ} |\psi_k(\mathbf{r},t)|^2$$

downfolding of equation of motion for extended orbitals (in region L-C-R) onto equation for orbital projected onto central region only but under influence of coupling to leads

TDDFT for transport

Equation of motion for orbital projected on central region

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

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

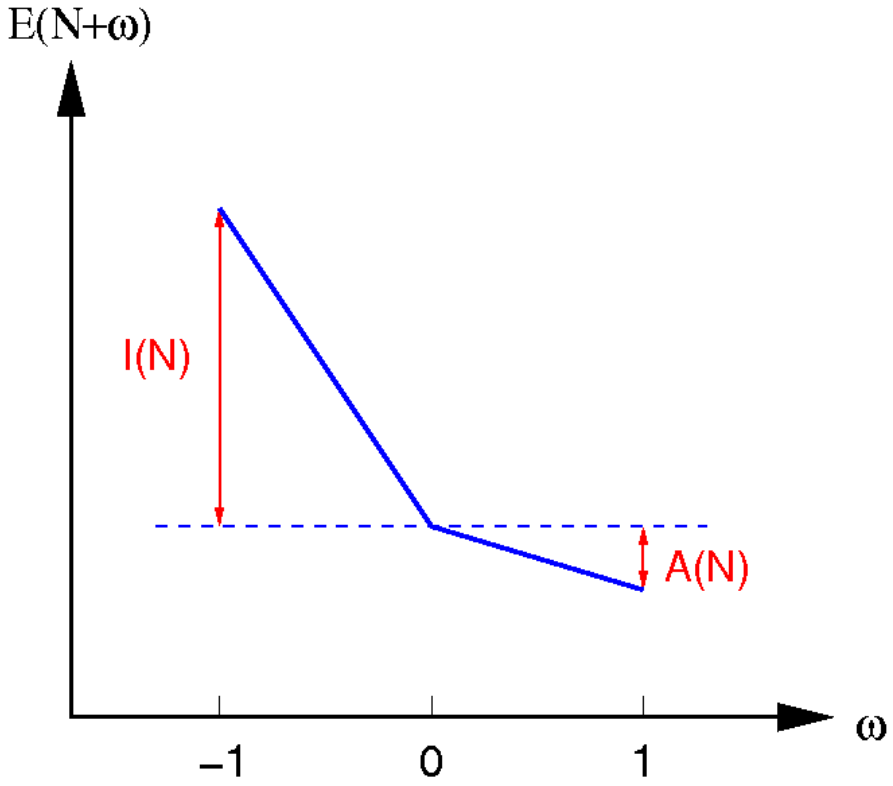
where (retarded) embedding self energy Σ^R_{emb} and (retarded) Green function g^R_α for isolated lead α describe coupling to leads

details in:

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

Derivative discontinuity in static DFT

total energy as function of (fractional) particle number is a series of straight lines (Perdew et al, PRL 49, 1691 (1982))



derivative discontinuity

$$\Delta = I(N) - A(N)$$

$I(N)$: ionization potential
$A(N)$: electron affinity
N : integer number of electrons

Derivative discontinuity in static DFT (cont.)

for given external potential $v(\mathbf{r})$, extend HK ground state energy functional to non-integer particle numbers:

derivative discontinuity

$$\Delta = \lim_{\omega \to 0} \left(\left. \frac{\delta E_v[n]}{\delta n(\mathbf{r})} \right|_{N+\omega} - \left. \frac{\delta E_v[n]}{\delta n(\mathbf{r})} \right|_{N-\omega} \right) = \Delta_{KS} + \Delta_{xc}$$

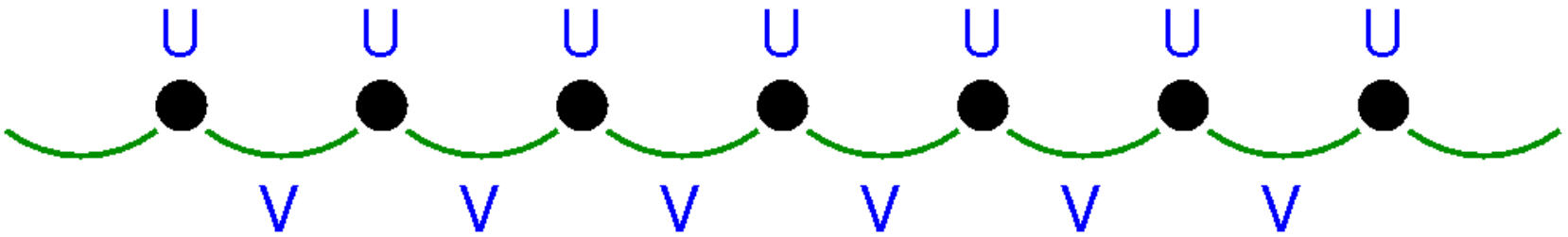
KS discontinuity $\Delta_{KS} = \varepsilon_{LUMO} - \varepsilon_{HOMO}$

xc contribution to discontinuity:

$$\Delta_{xc} = \lim_{\omega \to 0} \left(\left. \frac{\delta E_{xc}[n]}{\delta n(\mathbf{r})} \right|_{N+\omega} - \left. \frac{\delta E_{xc}[n]}{\delta n(\mathbf{r})} \right|_{N-\omega} \right)$$

note: for traditional functionals (LDA, GGA): $\Delta_{xc} = 0$!!

(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:

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

with parameter $\beta(U)$ 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 $n = 1$

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

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

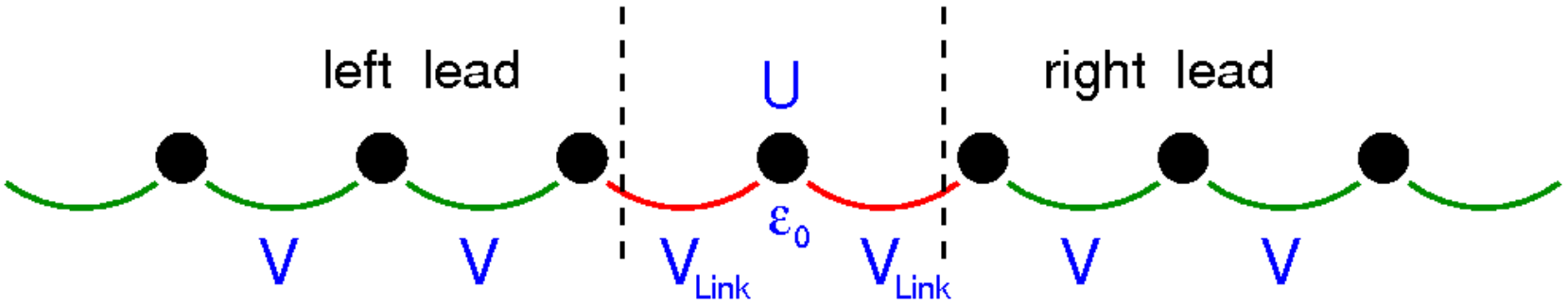
local approximation:

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 $v_{xc}(i,t) = v_{xc}^{BALDA}(n_i(t))$

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, on-site energy ε_0 at impurity

interested in case of weak links $|V_{\mathrm{Link}}| < |V| \longrightarrow$ use U/V_{link} as parameter in BALDA $\longrightarrow$ modified discontinuity at impurity

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

Self-consistency condition for steady state density

Landauer approach:
assume for biased system there exists steady state with density n
at impurity $\longrightarrow$ self-consistency condition for n

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

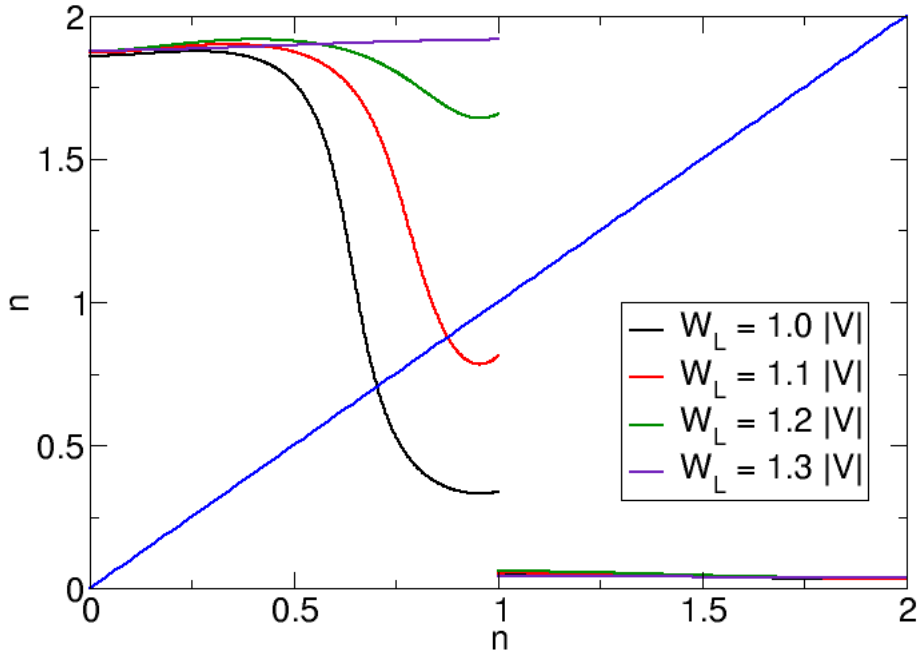
$$G(\omega) = [\omega - v_{KS}(n) - \Sigma_L(\omega - W_L) - \Sigma_R(\omega - W_R)]^{-1}$$

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

W_α: bias in lead α
$\Sigma_\alpha(\omega)$: embedding self energy for lead α

Self-consistency condition for steady state density

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

Smoothened discontinuity

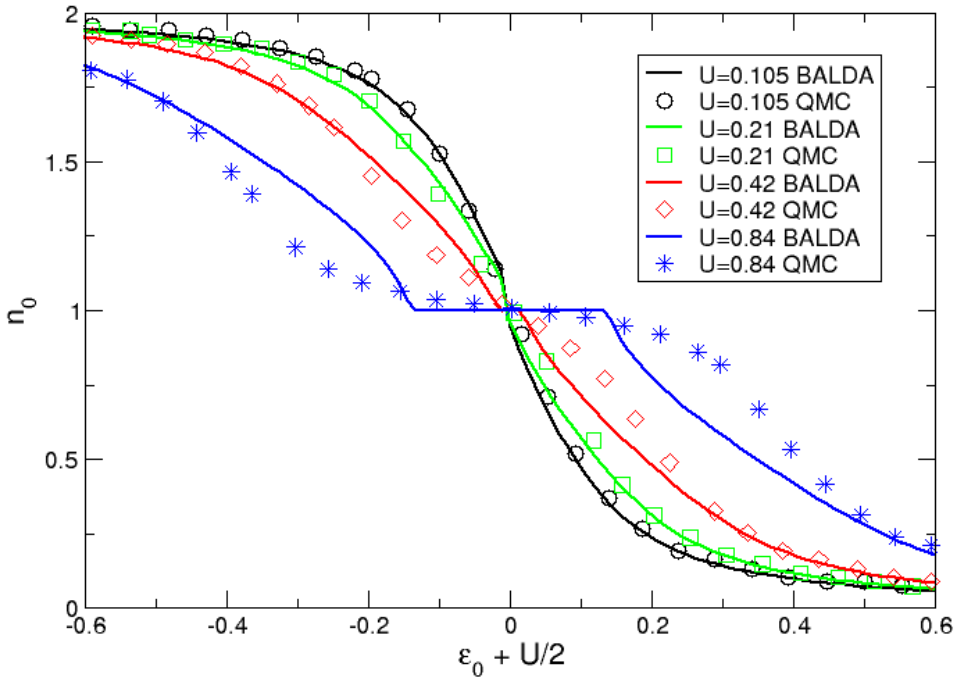
smoothen discontinuity

$$\tilde{v}_{xc}(n) = f(n)v_{xc}^{<}(n) + (1 - f(n))v_{xc}^{>}(n)$$
$$f(n) = [\exp((n-1)/a) + 1]^{-1}$$

where $v_{xc}^{<}(n)$ and $v_{xc}^{>}(n)$ are the functional relations for the BALDA xc potential for $n < 1$ and $n > 1$, respectively,
and a is a smoothening parameter
$\longrightarrow$ steady-state self-consistency condition always has solution!

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; $V_{\mathrm{link}} = 0.18$

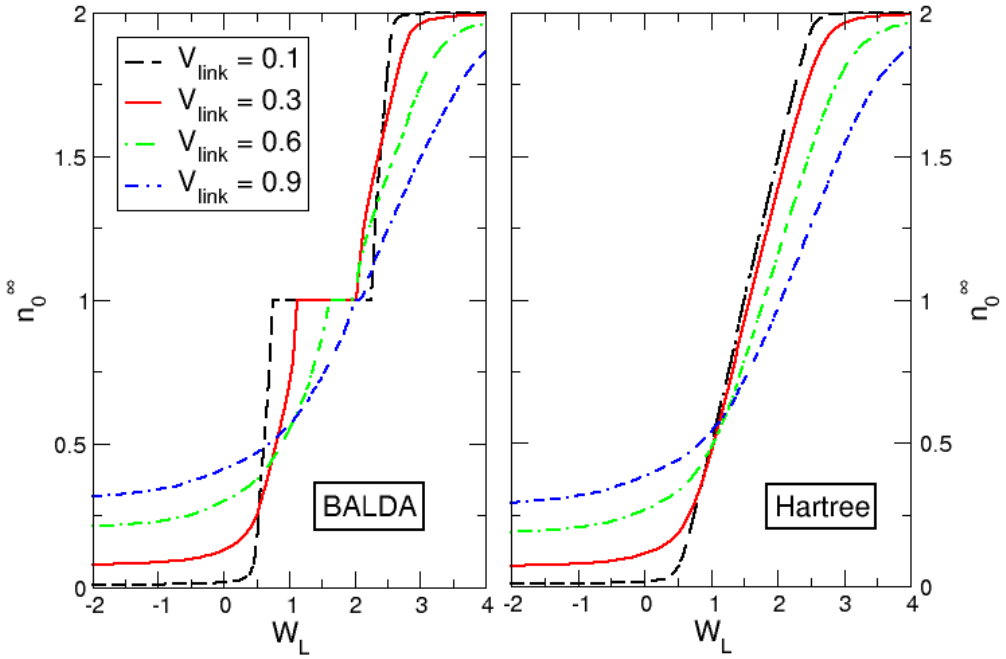


QMC results from:
X. Wang et al,
PRB **77**, 045119 (2008)

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}



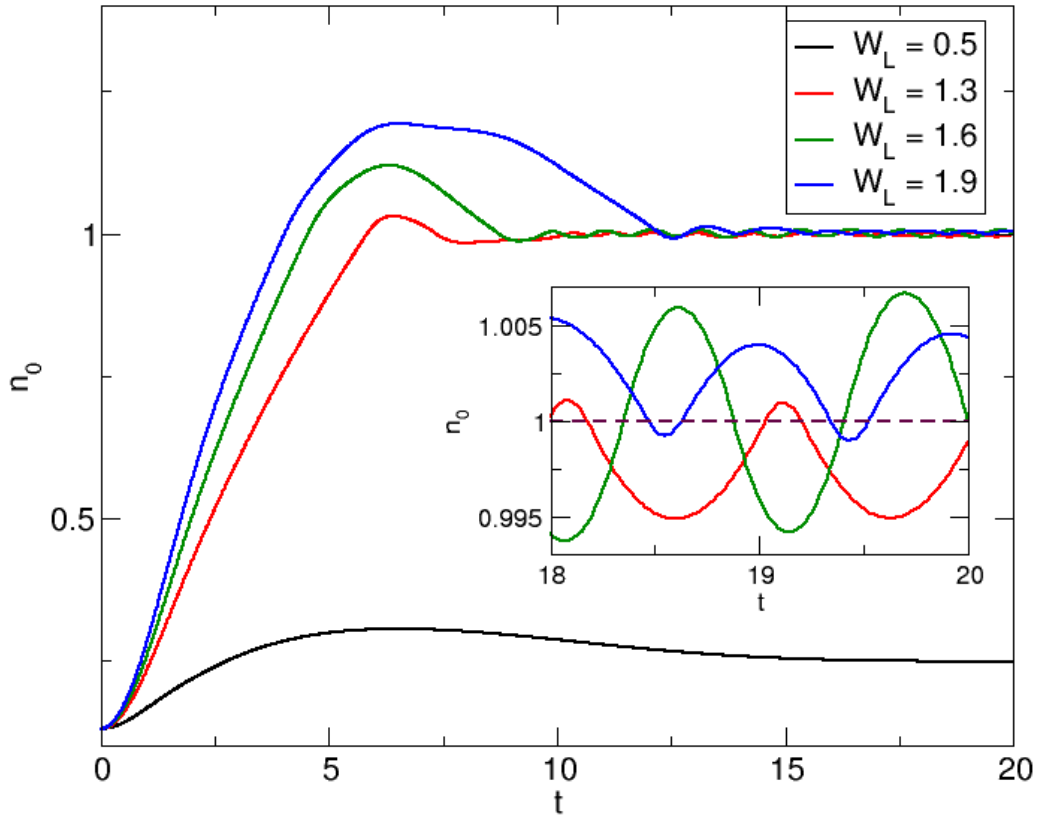
BALDA:
step structure
for small V_{link}
width of step: U
$\longrightarrow$ Coulomb blockade

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)

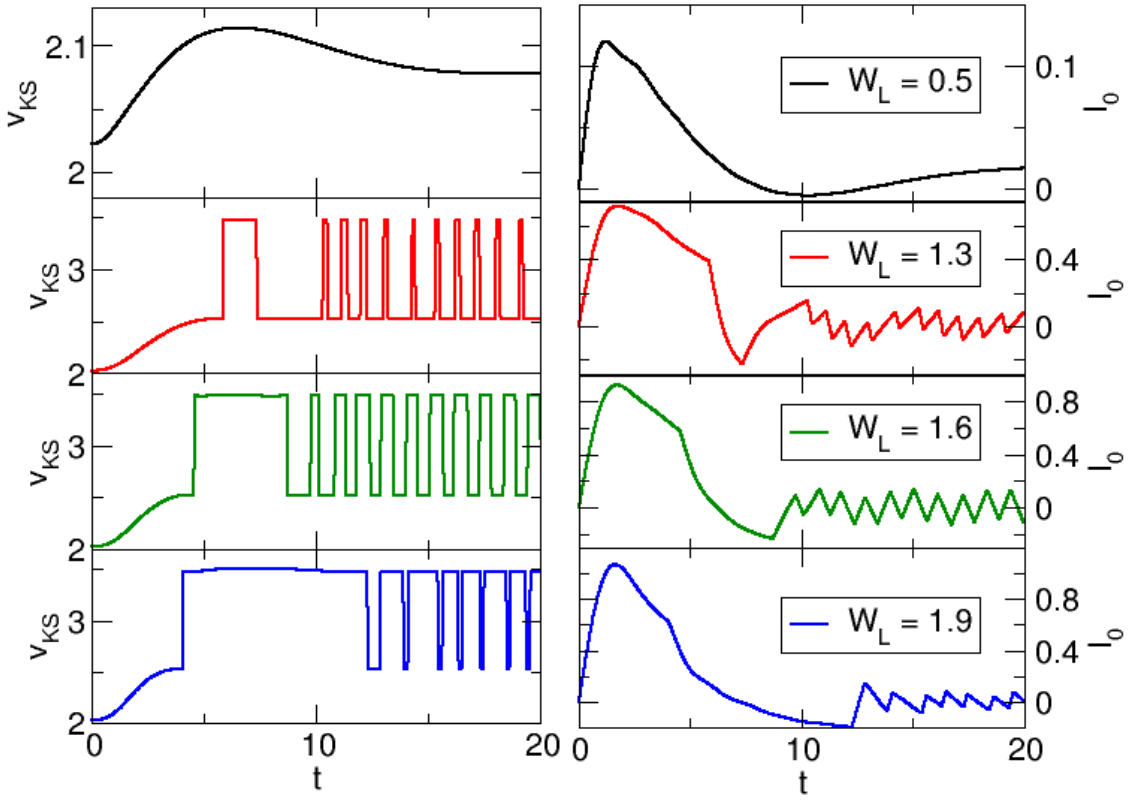
Time-dependent density in presence of discontinuity

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



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;

Are the oscillations real?

Two drastic approximations: local and adiabatic

- what to expect when one drops local, but keeps adiabatic approximation?
  steady-state condition will become a series of coupled, nonlinear equations which may have no solution if potential is discontinuous

- what to expect when one also drops adiabatic approximation, are oscillations destroyed?
  no idea, but if oscillations are killed we need much better functionals!

Summary

- TDDFT approach to transport
- Derivative discontinuity in transport crucial to describe Coulomb blockade
- absence of steady state in CB regime
  instead: TD picture of CB as dynamical state of charging and discharging of weakly coupled system

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)