Dynamical Coulomb blockade and the derivative discontinuity of time-dependent density functional theory

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Outline

- Time-dependent density functional theory for transport
- Derivative discontinuity and time-dependent picture of Coulomb blockade
- Summary
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

Outline
- Time-Dependent Density Functional Theory for Transport
  - Coulomb blockade and the role of the derivative discontinuity
  - Summary

TDDFT for transport

A simple impurity model for transport
(Static) DFT for the Hubbard model

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

\[
\hat{H}(t) = \begin{pmatrix}
H_{LL}(t) & H_{LC} & 0 \\
H_{CL} & H_{CC}(t) & H_{CR} \\
0 & H_{RC} & H_{RR}(t)
\end{pmatrix}
\]
TDDFT for transport

downfolding of equation of motion for extended orbitals (in region L-C-R) onto equation for orbital projected onto central region only

Equation of motion for orbital projected on central region

\[
\begin{align*}
[i \partial_t - \hat{H}_{CC}(t)]\psi_{k,C}(t) = & \int_0^t dt' \Sigma^{R}_{emb}(t, t')\psi_{k,C}(t') + \sum_{\alpha} H_{C}\alpha g^{R}_{\alpha}(t, 0)\psi_{k,\alpha}(0)
\end{align*}
\]

where (retarded) embedding self energy $\Sigma^{R}_{emb}$ and (retarded) Green function $g^{R}_{\alpha}$ for isolated lead $\alpha$ describe coupling to leads

details in:
Simple impurity model for transport

One interacting impurity, Hubbard-like on-site interaction $U$, non-interacting leads, hopping $V$ in leads and hopping $V_{\text{Link}}$ from leads to impurity, on-site energy $\varepsilon_0$ at impurity

At time $t = 0$, switch on bias $W_\alpha$ in lead $\alpha$ and follow time evolution

In TDDFT: need exchange-correlation potential
(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 \( e^{BA}_{xc}(n, U) \) from this parametrization
(Static) DFT for the Hubbard model

**derivative discontinuity at** \( n = 1 \)

\[
\Delta_{xc} = \lim_{\epsilon \to 0^+} \left[ v_{xc}^{BALDA}(n = 1 + \epsilon) - v_{xc}^{BALDA}(n = 1 - \epsilon) \right] \\
= U - 4|V| \cos \left( \frac{\pi}{\beta(U)} \right)
\]

**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

\[
v_{xc}(i, t) = v_{xc}^{BALDA}(n_i(t))
\]
TD density and KS potential in presence of discontinuity

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

density shows small oscillations around integer occupation

for some parameters: system does not evolve towards a steady but towards a dynamical state

TD KS potential: series of almost rectangular potential steps
Self-consistency condition for steady state density

Landauer approach:
assume there exists steady state with density $n^\infty$ at impurity
$
\rightarrow$ self-consistency condition for $n^\infty$

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

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

$$
v_{KS}(n) = \varepsilon_0 + \frac{1}{2} Un + v_{xc}^{BALDA}(n) 
$$
Self-consistency condition for steady state density

l.h.s. and r.h.s. of self-consistency condition for $n^\infty$

![Graph showing the self-consistency condition for steady state density](image)

no solution for steady state density for some values of the bias, exactly those values for which TD approach gives dynamical state!!

to understand physics of this regime $\rightarrow$ smoothen xc discontinuity
Smoothened discontinuity: steady-state density vs. bias

steady-state density as function of bias for different hoppings from lead to impurity

step structure for small $V_{\text{Link}}$ width of step: $U$

→ Coulomb blockade

note: crucial role of discontinuity

the role of the discontinuity in steady-state transport has also been discussed in C. Toher et al, PRL 95, 146402 (2005)
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:
see also: C.A. Ullrich, Physics Viewpoint 3, 47 (2010)