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

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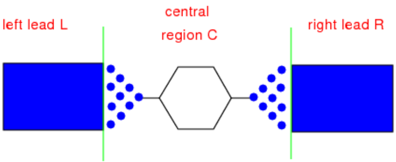
Outline

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

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

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

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

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

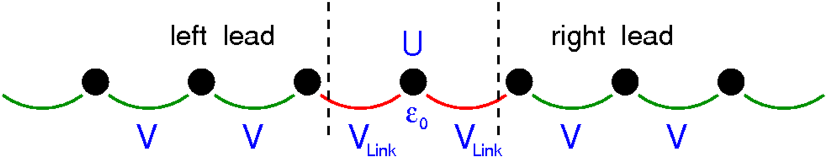
$$\int_0^t \mathrm{d}\bar{t}\ \Sigma^R_{emb}(t,\bar{t})\psi_{k,C}(\bar{t}) + \sum_\alpha H_{C\alpha} g^R_\alpha(t,0)\psi_{k,\alpha}(0)$$

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)

Simple impurity model for transport

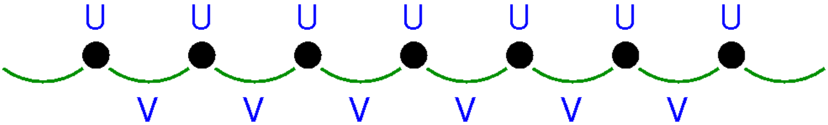


one interacting impurity, Hubbard-like on-site interaction U, non-interacting leads, hopping V in leads and hopping $V_{\rm Link}$ from leads to impurity, on-site energy ε_0 at impurity

at time $t = 0$, switch on bias W_α in lead α 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 $e_{xc}^{BA}(n,U)$ 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:

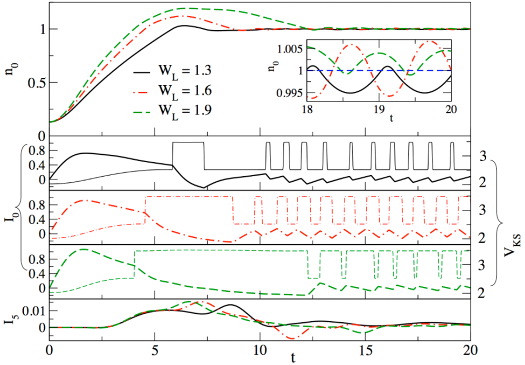
for non-uniform Hubbard models, i.e., non-constant on-site energies or even different interactions at each site:
use $e_{xc}^{BA}(n_i, U_i)$ as xc energy at site i (Bethe ansatz LDA, BALDA)

adiabatic approximation:

time-dependence of TDDFT xc potential at site i through
$v_{xc}(i,t) = v_{xc}^{BALDA}(n_i(t))$

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 $V_{\rm Link} = 0.3V$



density shows small oscillations around integer occupation

TD KS potential: series of almost rectangular potential steps

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

Self-consistency condition for steady state density

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

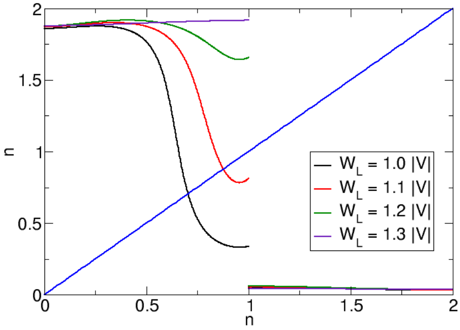
$$n^\infty = 2 \sum_{\alpha=L,R} \int_{-\infty}^{\varepsilon_f + W_\alpha} \frac{\mathrm{d}\omega}{2\pi} \Gamma(\omega - W_\alpha) |G(\omega)|^2$$

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

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

Self-consistency condition for steady state density

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

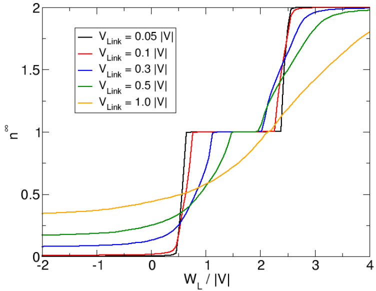


no solution for steady state density for some values of the bias, exactly those values for which TD approach gives dynamical state !!

to understand physics of this regime $\longrightarrow$ 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_{Link}
width of step: U
$\longrightarrow$ Coulomb blockade

<u>note:</u> 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:

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)