The derivative discontinuity in transport

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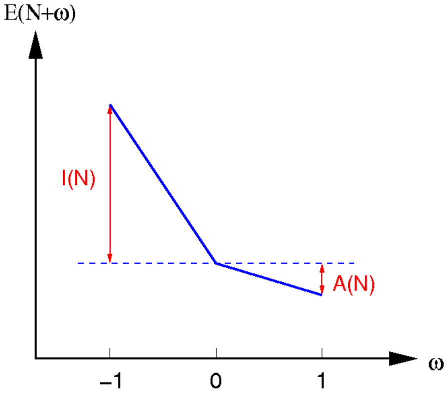
Outline

- Setting the stage
- Derivative discontinuity and time-dependent picture of Coulomb blockade
- The Kondo effect: what TDDFT has to say
- Summary

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

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

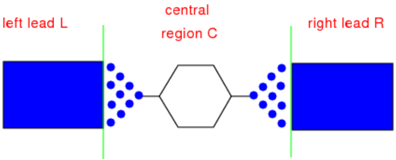
KS discontinuity $\Delta_{KS} = \varepsilon_{LUMO} - \varepsilon_{HOMO}$

xc contribution to discontinuity:

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

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

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

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

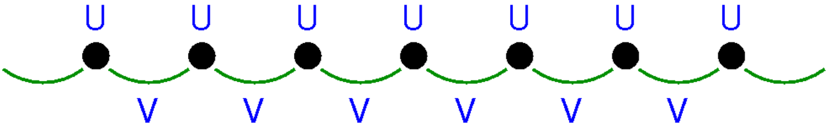
$$\int_0^t \mathrm{d}\bar{t}\, \Sigma^R_{emb}(t,\bar{t})\psi_{k,C}(\bar{t}) + \sum_\alpha H_{C\alpha} g^R_\alpha(t,0)\psi_{k,\alpha}(0)$$

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)

(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 $n = 1$

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

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

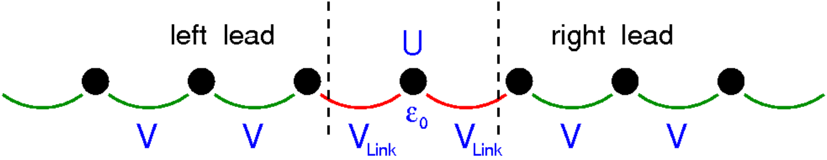
local approximation:

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 $V_{\rm Link}$ from leads to impurity, on-site energy ε_0 at impurity

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

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

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

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

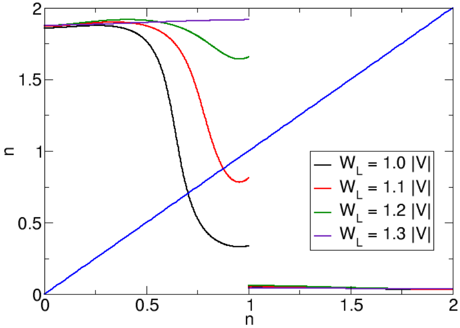
$$G(\omega) = [\omega - v_{KS}(n) - \Sigma_L(\omega - W_L) - \Sigma_R(\omega - W_R)]^{-1}$$

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

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

Steady state self-consistent density for impurity model

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

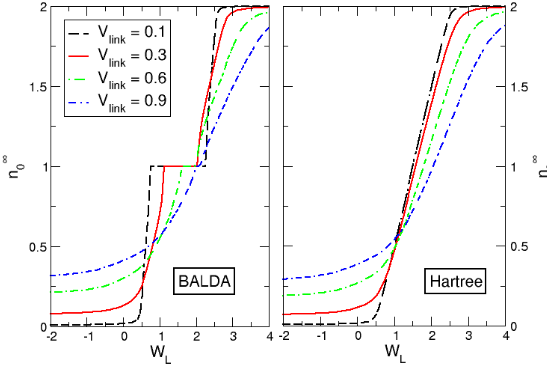


no solution for steady state density for some values of the bias.

to understand physics of this regime ⟶ smoothen xc discontinuity

Steady-state density vs. bias

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



<u>BALDA:</u>

step structure for small V_{link}

width of step: U

$\rightarrow$ Coulomb blockade

<u>Hartree:</u>

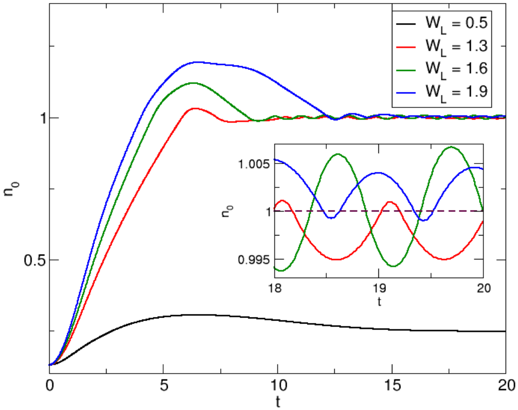
no step structure

$\rightarrow$ crucial role of discontinuity

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

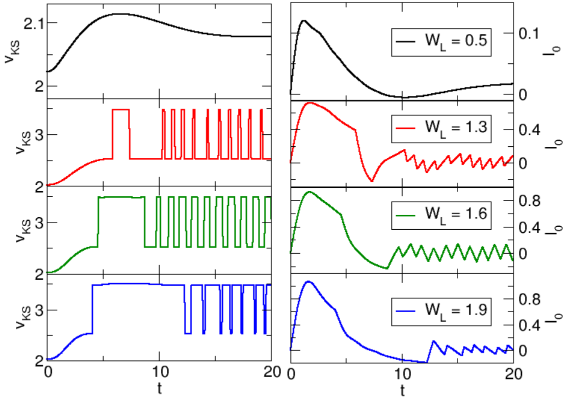
Time-dependent density in presence of discontinuity

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

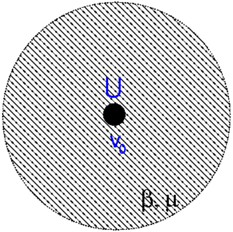
Time-dependent KS potentials and currents



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

Ref: S. Kurth, G. Stefanucci, E. Khosravi, C. Verdozzi, E.K.U. Gross, PRL **104**, 236801 (2010)

Single-site model to construct finite temperature functional



4 states in Fock space: $|0\rangle$, $|\uparrow\rangle$, $|\downarrow\rangle$, $|\uparrow\downarrow\rangle$

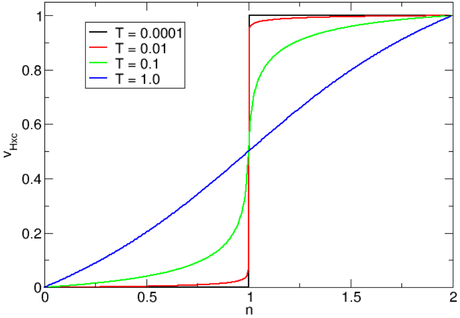
calculate density $n(v_0 - \mu) = n(\tilde{v}_0)$
invert analytically $\longrightarrow \tilde{v}_0(n)$

non-interacting KS system: density $n_s(\tilde{v}_s)$
invert analytically $\longrightarrow \tilde{v}_s(n_s)$

Hartree-xc potential: $$v_{Hxc}(n) = \tilde{v}_s(n) - \tilde{v}_0(n)$$

Hartree-xc potential for single-site model

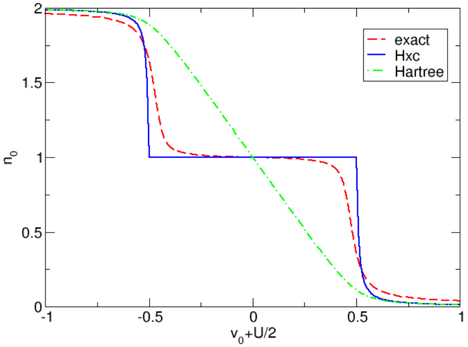
Hartree-xc potential for different temperatures (in units of U)



derivative discontinuity emerges naturally in the zero-temperature limit

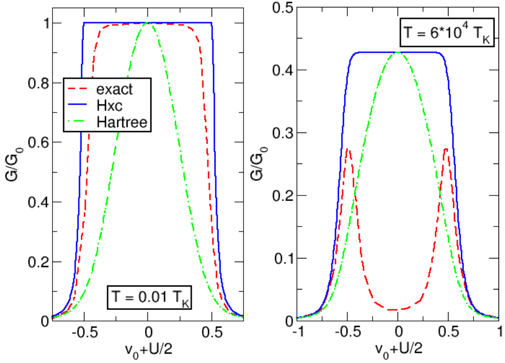
Density at zero temperature compared to exact results

local approximation: use single-site potential on impurity, vanishing KS potential in leads, should be reasonable approximation for weak coupling



$U/\gamma = 100$; γ: DOS in wide-band limit; density pinned to 1 due to discontinuity;

Finite temperature conductance with Landauer



$T << T_K$: Kondo plateau in conductance due to discontinuity;

$T >> T_K$: plateau *not* destroyed; no Coulomb blockade peaks

exact data from: Izumida, Sakai, J. Phys. Soc. Jpn., 2005

Finite temperature conductance with Landauer

two ways to understand $T = 0$ result:

- Meir-Wingreen formula for conductance:

$$\frac{G}{G_0} = \gamma^2 |\mathcal{G}(\mu)|^2 \frac{\gamma - \mathrm{Im}\,\Sigma(\mu)}{\gamma}$$

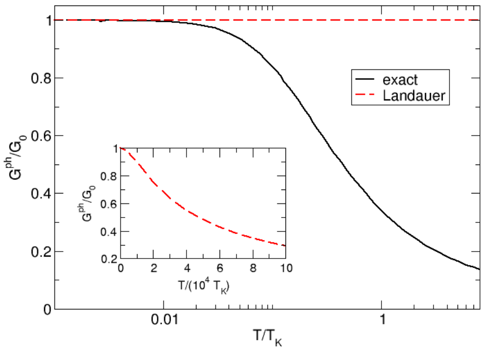
  with many-body GF $\mathcal{G}(\omega)$ at impurity and self energy Σ at Fermi energy: $\mathrm{Im}\,\Sigma(\mu) = 0 \longrightarrow$ can describe conductance by a KS potential $v_s = v_0 + \mathrm{Re}\Sigma(\mu)$

- Friedel sum rule (Langreth):

  conductance determined by density n_0 on dot: $G = G(n_0)$
  if KS potential gives good density $\longrightarrow$ good conductance

Finite temperature conductance with Landauer

note: at particle-hole symmetric point $v_0 = -U/2$ our approximation gives *exact* KS potential for all temperatures



for finite T: Landauer *does not* give correct conductance although static KS potential is exact!

exact results from T.A. Costi, PRL (2000)

Dynamical xc corrections beyond Landauer

TDDFT formula for current to linear order in bias:

$$I = -G_0(V_L + V_{L,xc} - V_R - V_{R,xc}) \int \mathrm{d}\omega \frac{\partial f_\beta(\omega)}{\partial \omega} \mathcal{T}(\omega)$$

with transmission function $\mathcal{T}(\omega)$ and dynamical xc correction

$$V_{\alpha,xc} = \lim_{i\to\infty} \sum_j f_{xc}(i\alpha, j)\delta n_j$$

$\longrightarrow$ explicit example that xc correction to conductance can be necessary and even exact (static) KS potential is not enough!!

A fashionable thing to do...

- Bergfield, Liu, Burke, Stafford, "Kondo effect given exactly by density functional theory", arXiv:1106.3104, June 17, 2011
- Tröster, Schmitteckert, Evers, "DFT-based transport calculations: Friedel's sum rule and the Kondo effect", arXiv:1106.3669, June 21, 2011
- Stefanucci, Kurth, "Towards a description of the Kondo effect using time-dependent density functional theory", arXiv:1106.3728, June 21, 2011

Collaborators:

On dynamical Coulomb blockade:

- G. Stefanucci, Univ. Tor Vergata, Rome, Italy
- E. Khosravi and E.K.U. Gross, MPI Halle, Germany
- C. Verdozzi, Univ. Lund, Sweden

On Kondo:

- G. Stefanucci, Univ. Tor Vergata, Rome, Italy

Summary

- Derivative discontinuity in transport
- absence of steady state in CB regime; instead: TD picture of CB as dynamical state of charging and discharging of weakly coupled system
- conductance plateau as function of gate (Kondo) at $T = 0$
- understand in terms of Meir-Wingreen formula and Friedel sum rule
- finite T: Landauer not enough; TDDFT dynamical xc corrections essential

If you want to describe strongly correlated systems, the derivative discontinuity is your friend!