Multiple steady states and derivative discontinuities in time-dependent transport

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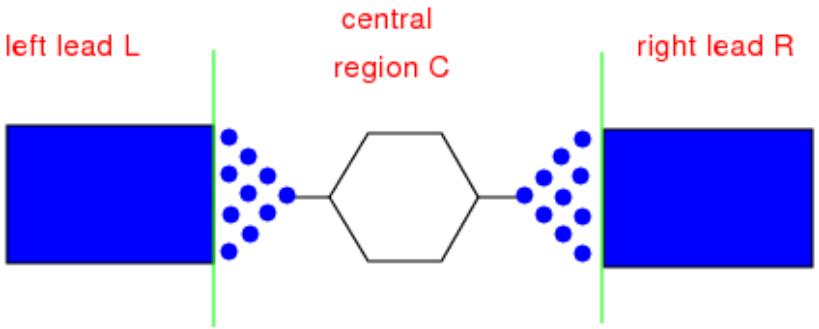
Outline

- Time-dependent transport: Why and how?
- Real-time switching between multiple steady states
- 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
- the steady state is achieved at the end of an evolution process
- can describe TD phenomena: transients, TD bias, external TD fields, ...
- methods:
  - TDDFT: in principle exact (unlike standard DFT+NEGF approach)
  - MBPT: Kadanoff-Baym equation of motion for nonequlibrium (Keldysh) GF

Time-Dependent Density Functional Theory



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

details in:

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

Time-Dependent Density Functional Theory

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

$$\Sigma^R_{emb}(t,t') = \sum_{\alpha=L,R} H_{C\alpha} g^R_\alpha(t,t') H_{\alpha C}$$

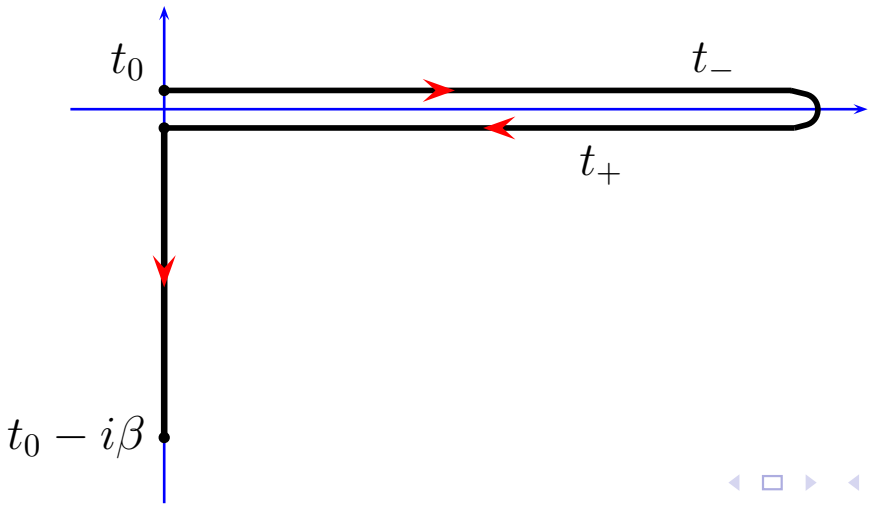
and (retarded) Green function g^R_α for isolated lead α

Kadanoff-Baym Equations

Keldysh Green function

$$G(1,2) = -i\frac{Tr\{\hat{U}(t_0 - i\beta, t_0)\mathcal{T}_C[\hat{\psi}_H(1)\hat{\psi}_H^\dagger(2)]\}}{Tr\{\hat{U}(t_0 - i\beta, t_0)\}}$$

β: inverse temperature, $\hat{\psi}_H(1)$: Heisenberg field operator
$\hat{U}$: time evolution operator, $\mathcal{T}_C$: time-ordering operator on Keldysh contour



Kadanoff-Baym Equations (cont.)

Equation of motion for Keldysh GF

$$[i\partial_{t_1} - \hat{H}(1)]G(1,2) = \delta(1,2) + \int_{\mathcal{C}} \mathrm{d}3\, \Sigma^{MB}[G](1,3)G(3,2)$$

$\Sigma^{MB}[G](1,2)$: many-body self energy (functional of G), assume it is only non-vanishing in central region and depends only on central block of GF

$$\Sigma^{MB} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & \Sigma^{MB}_{CC}[G_{CC}] & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Kadanoff-Baym Equations (cont.)

Equation of motion for Keldysh GF in central region only

$$[i\partial_t - \hat{H}_{CC}(t)]G_{CC}(t,t') = \delta(t-t')$$

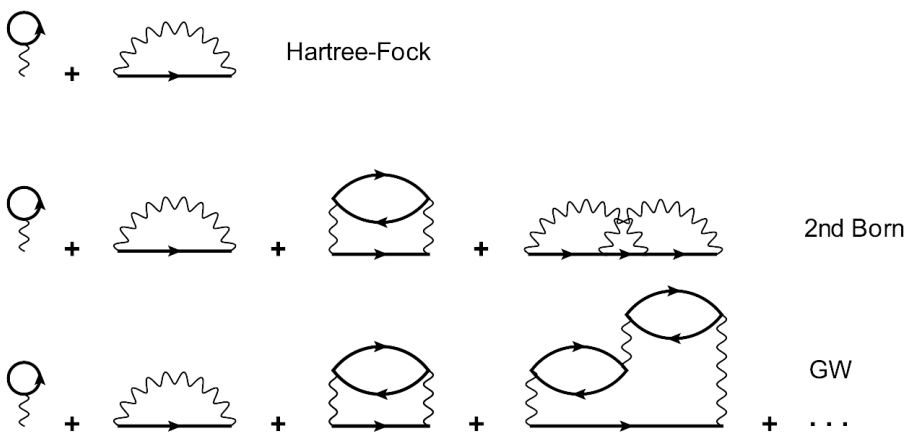
$$+ \int_{\mathcal{C}} \mathrm{d}\bar{t}\, [\Sigma_{emb}(t,\bar{t}) + \Sigma^{MB}_{CC}[G_{CC}](t,\bar{t})]G(\bar{t},t')$$

with embedding self energy $\Sigma_{emb}(t,t')$ as before

Density from lesser Green function

$$n(\mathbf{r},t) = -iG^{<}(\mathbf{r},t,\mathbf{r},t^{+})$$

approximations used for many-body self energy:



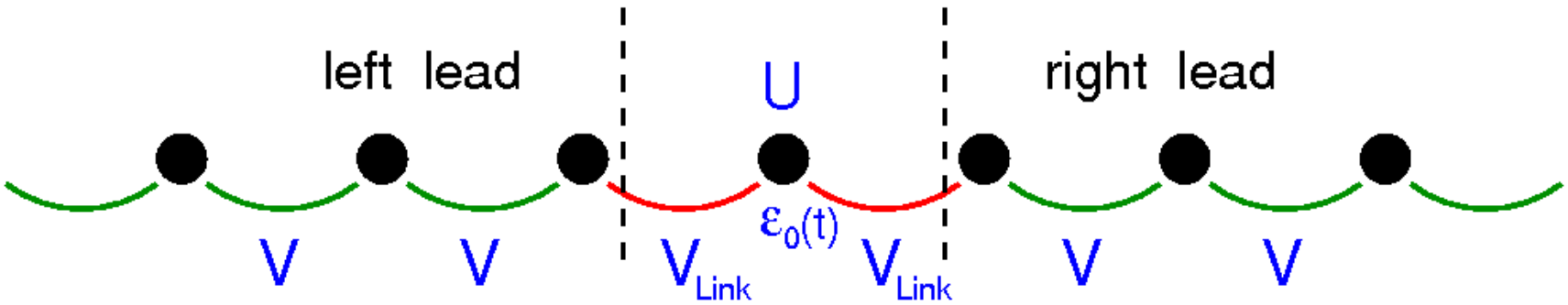
details in:

P. Myöhänen, A. Stan, G. Stefanucci, R. van Leeuwen, PRB **80**, 115107 (2009)

Multistabilities in TD Transport

- Simple impurity model
- Steady-state condition in DFT: multiple solutions
- Time-dependent switching between steady states
- Inclusion of correlation

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 U_α in lead $\alpha = L, R$, assume there exists steady state with density n at impurity
$\longrightarrow$ self-consistency condition for n

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

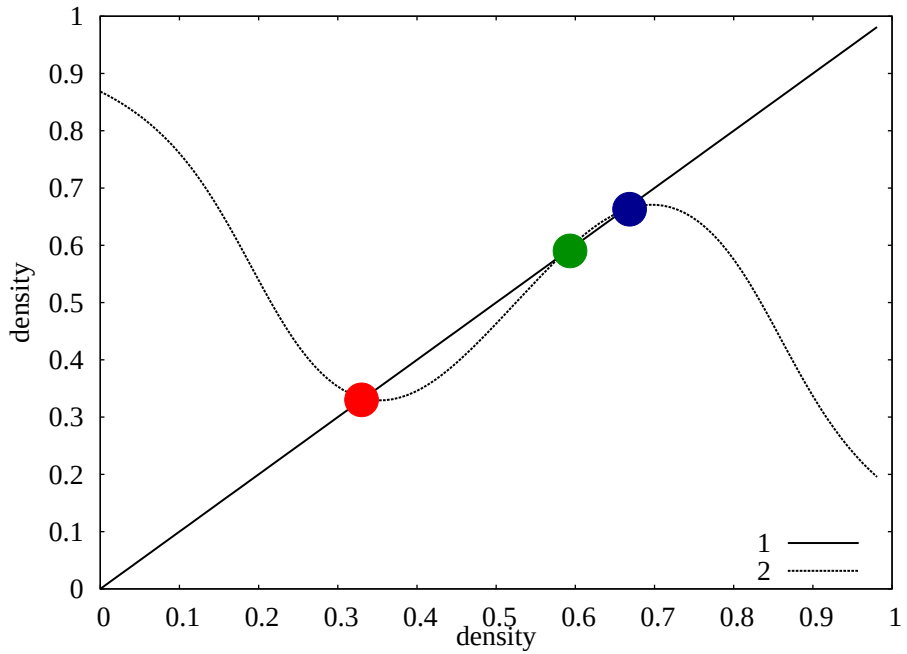
$$G(\omega) = [\omega - v_{KS}(n) - \Sigma(\omega - U_L) - \Sigma(\omega - U_R)]^{-1}$$

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

here: use Hartree (which, for our model, is equivalent to HF), i.e., $v_{xc} \equiv 0$

Multiple solutions for steady-state condition in DFT

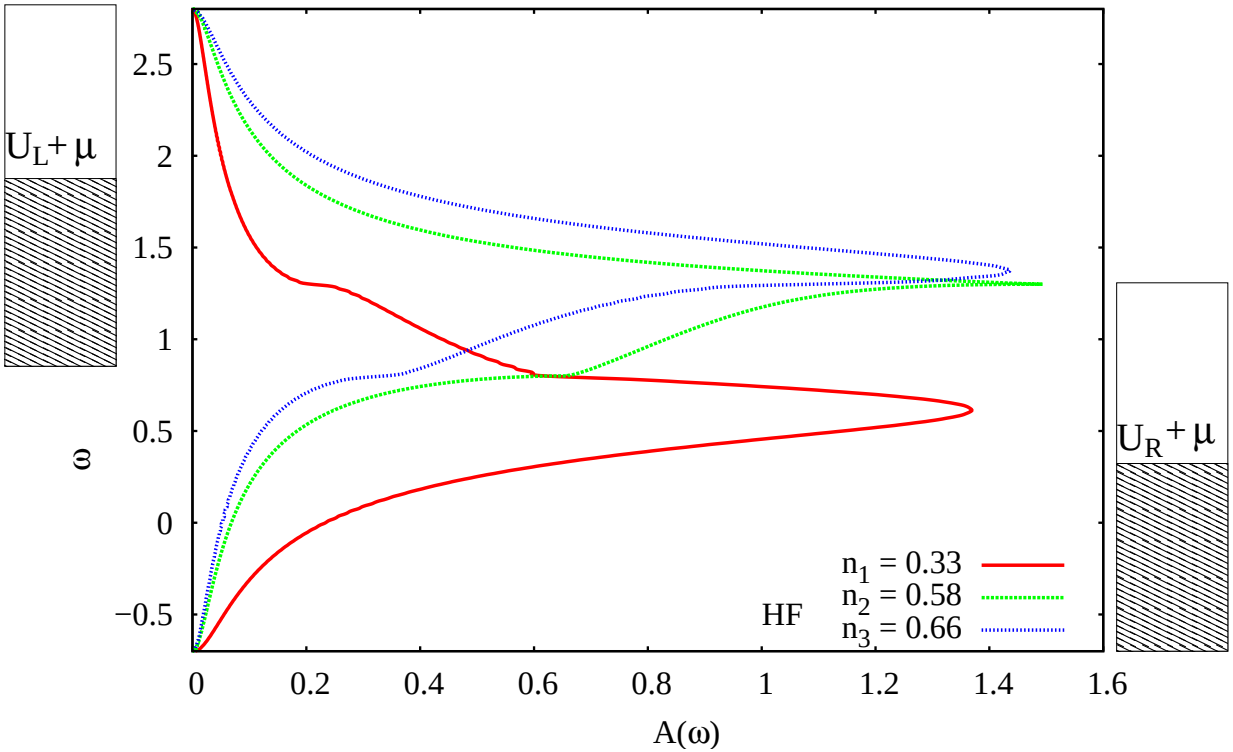
lead bands from $-2|V|$ to $2|V|$, Fermi energy $\varepsilon_F = 0.6|V|$, on-site interaction $U = 4|V|$, hopping to impurity $V_{\text{Link}} = 0.7V$, on-site energy $\varepsilon_0 = 0$, right bias $U_R = 0$, left bias $U_L = 3|V|$



three solutions for self-consistent density

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

Spectral functions for multiple steady-state solutions

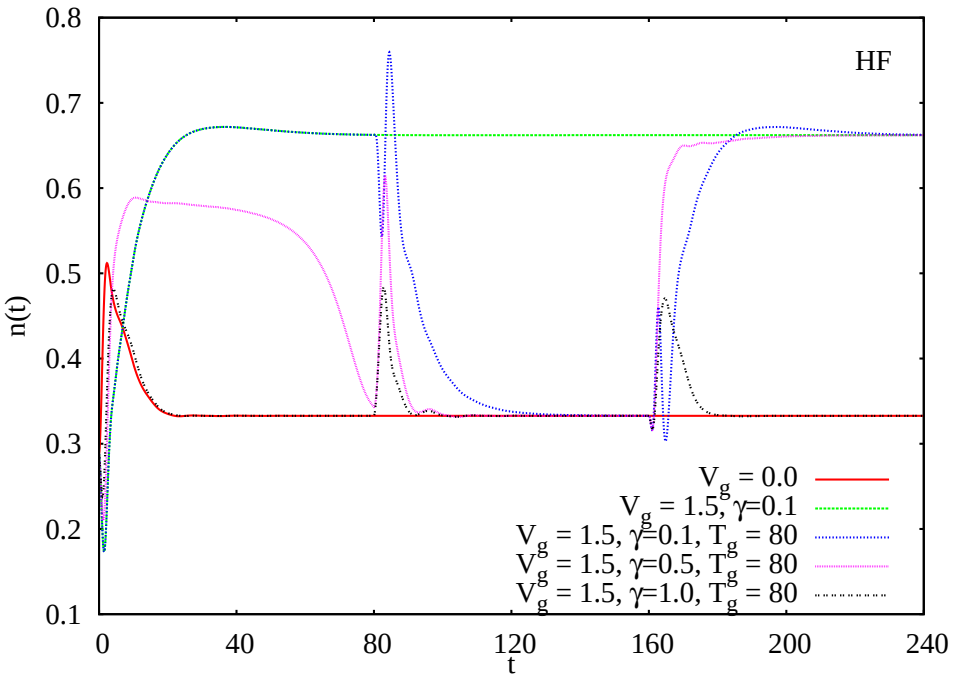


spectral function for n_1 peaked in energy range of right band, for n_3 peaked in range of left band, for n_2 peaked at top of right band

Time-dependent switching between different steady states

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

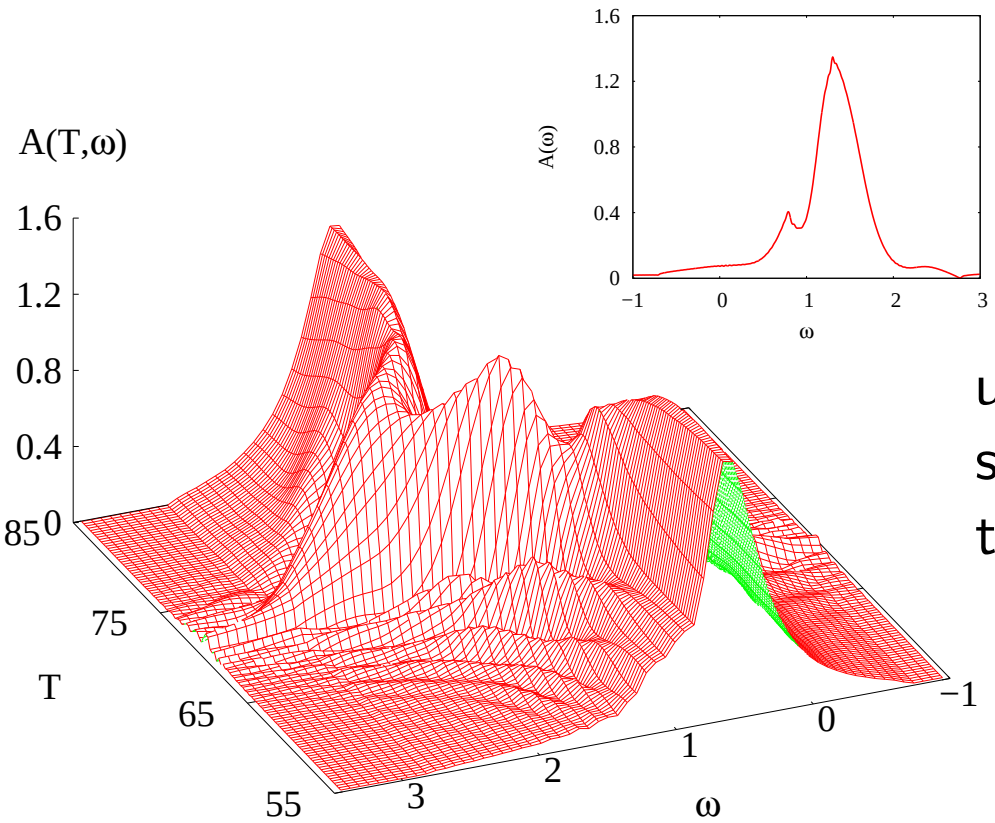
$$\varepsilon_0(t) = \begin{cases} V_g \exp(-\gamma t) & \text{if } 0 < t < T_g \\ -V_g \exp(-\gamma(t - T_g)) & \text{if } T_g < t < 2T_g \\ V_g \exp(-\gamma(t - 2T_g)) & \text{if } T_g < t < 2T_g \end{cases}$$



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

time-dependent sprectral function (from Kadanoff-Baym approach)

$$A(T,\omega) = \mathrm{Im}\mathrm{Tr}\int_{-2T}^{2T} \frac{\mathrm{d}\omega}{2\pi} \exp(i\omega\tau)\left(G^{>} - G^{<}\right)\left(T+\frac{\tau}{2}, T+\frac{\tau}{2}\right)$$

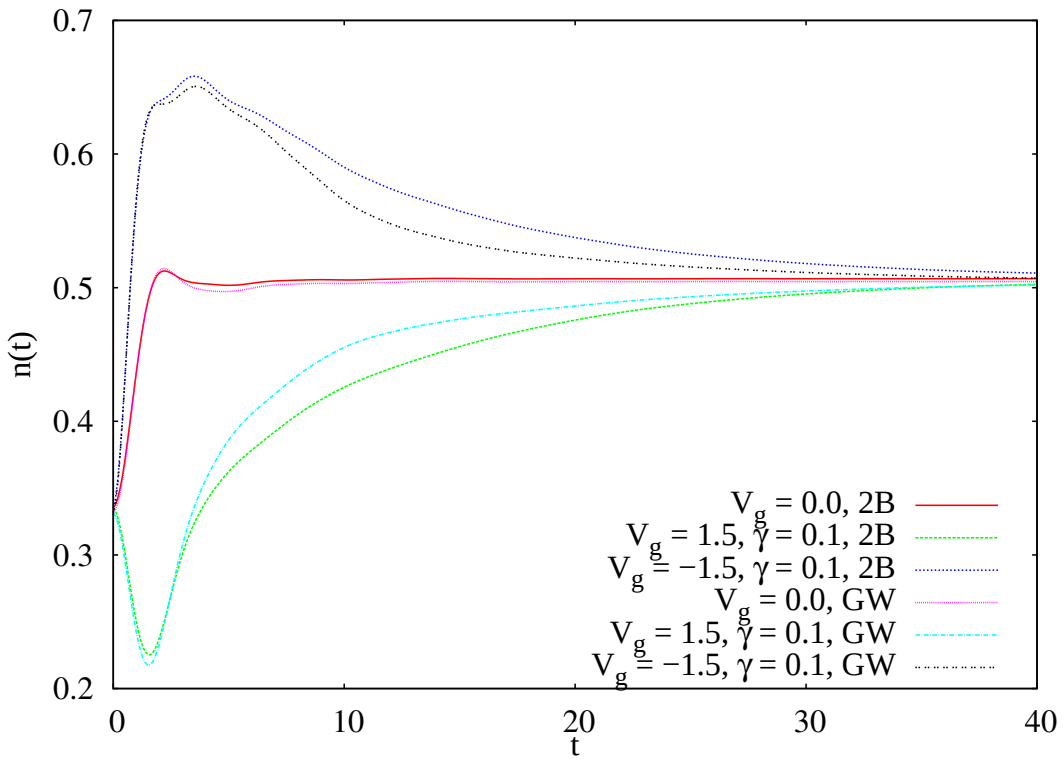


$A(T=70,\omega)$

unstable, second steady state appears during the transition !

Inclusion of correlation in GW and second Born

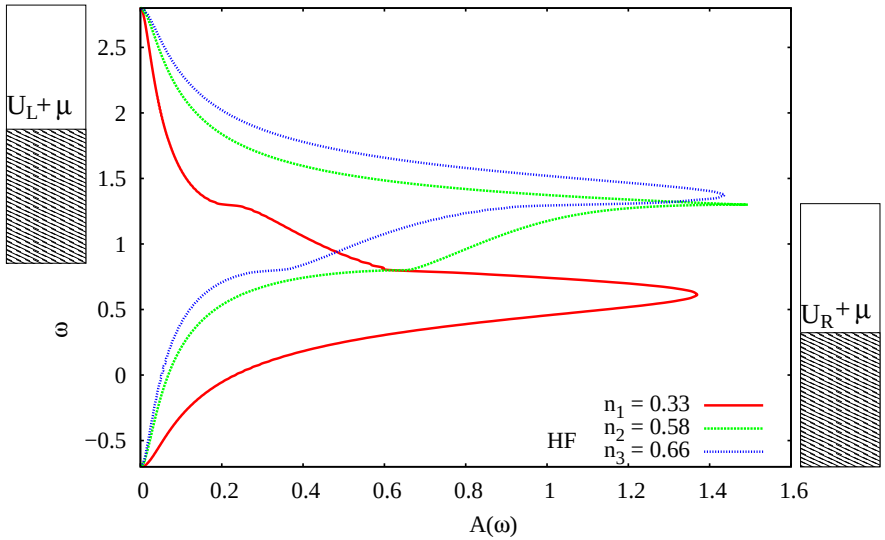
TD density for same parameters in GW and second Born



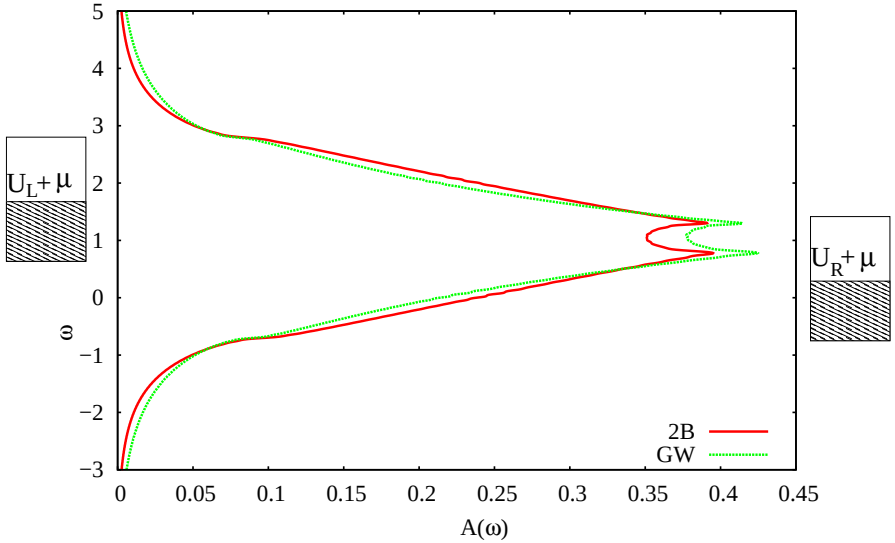
only one steady state is found $\longrightarrow$ correlation can destroy bistability

Spectral Functions in HF, 2B, and GW in steady state

HF



2B and GW



HF: spectral density of stable steady states well localized in one of the lead bands

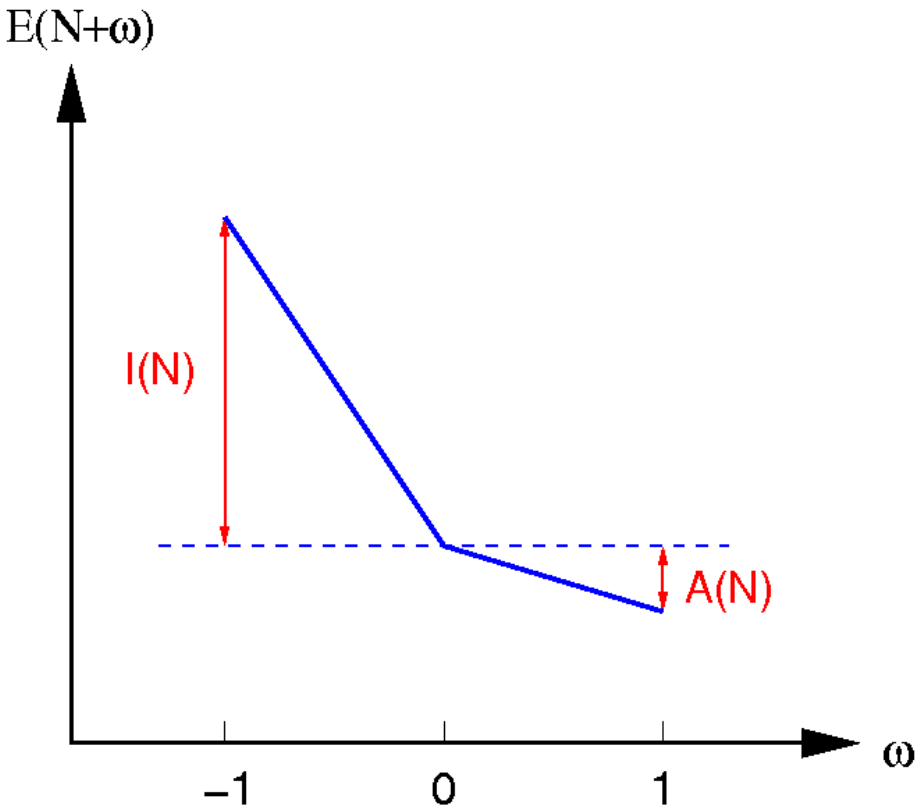
2B and GW: spectral density spread out over whole energy range

The Role of the Derivative Discontinuity in TD Transport

- Derivative discontinuity in static DFT
- Static DFT for the Hubbard model
- The simple model again
  - Steady-state condition without solution
  - Smoothen discontinuity: step structure for density vs. bias
  - Effect of discontinuity on TD density

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

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

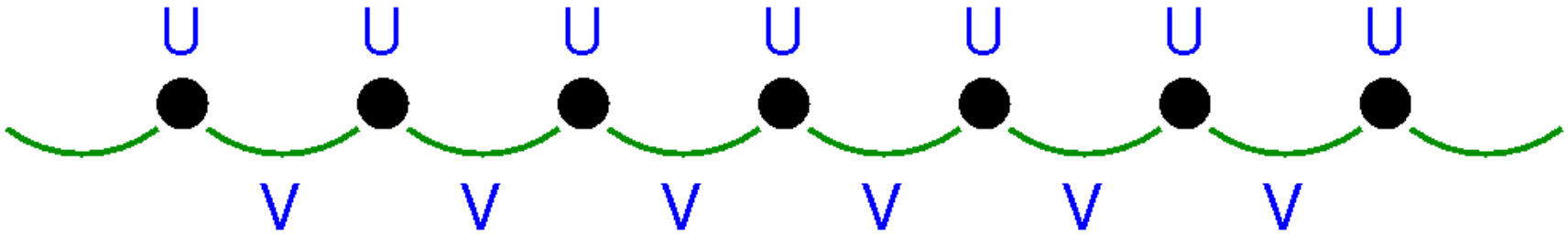
KS discontinuity $\Delta_{KS} = \varepsilon_{LUMO} - \varepsilon_{HOMO}$

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)):

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 β depending on interaction strength U
one can extract xc energy $e_{xc}^{BA}(n,U)$ from this parametrization

(Static) DFT for the Hubbard model

important property [N.A. Lima et al (EPL 60, 601 (2002))]:

derivative discontinuity at $n = 1$

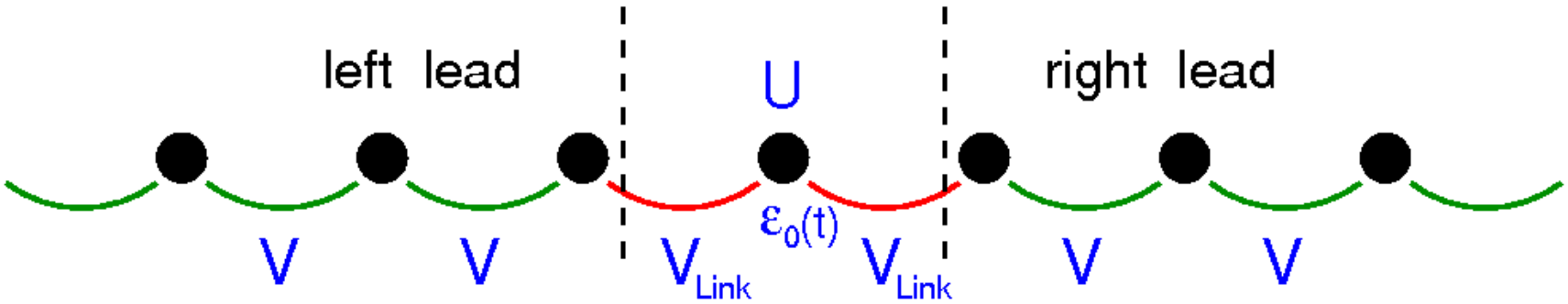
$$\Delta = \lim_{\epsilon \longrightarrow 0^+} [v_{xc}^{BALDA}(n = 1 + \epsilon) - v_{xc}^{BALDA}(n = 1 - \epsilon)]$$

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

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)

Simple impurity model again



model as before (except for discontinuous xc potential at impurity)
mainly interested in case of weak links $|V_{\text{Link}}| < |V|$

$\longrightarrow$ modified discontinuity at impurity

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

Self-consistency condition for steady state density

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

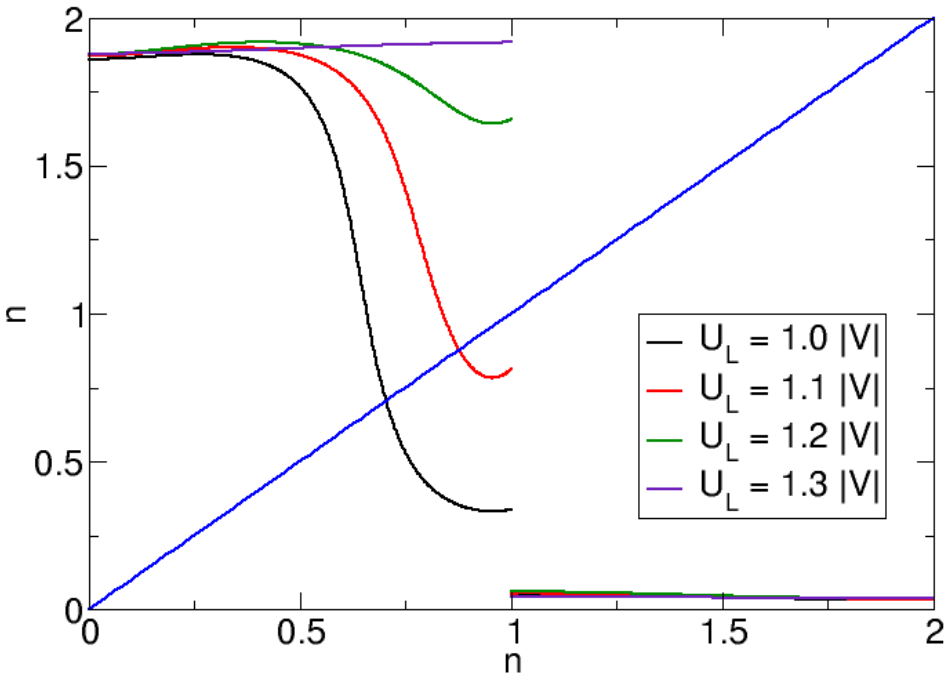
$$n = 2 \sum_{\alpha=L,R} \int_{-\infty}^{\varepsilon_f + U_\alpha} \frac{\mathrm{d}\omega}{2\pi} \Gamma(\omega - U_\alpha) |G(\omega)|^2$$

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

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

Self-consistency condition for steady state density

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



no solution for some bias values !!

Smoothened discontinuity

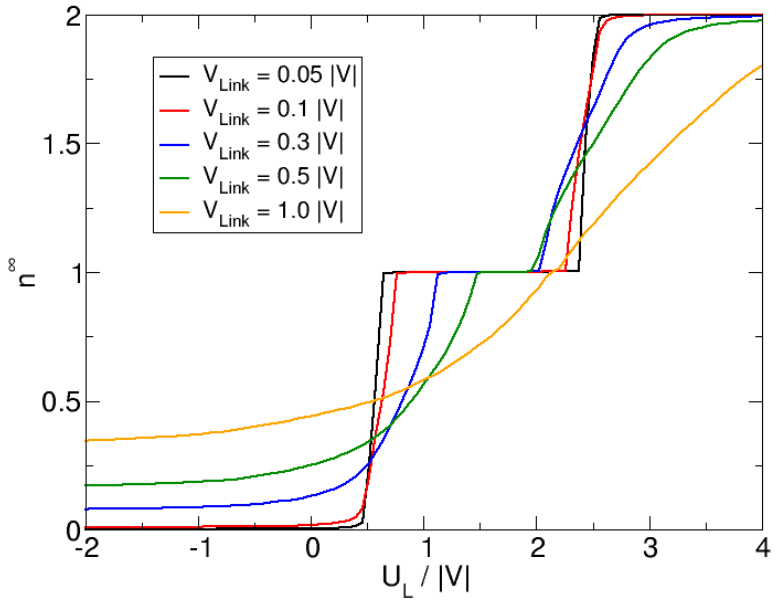
smoothen discontinuity

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

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!

Smoothened discontinuity: steady-state density vs. bias

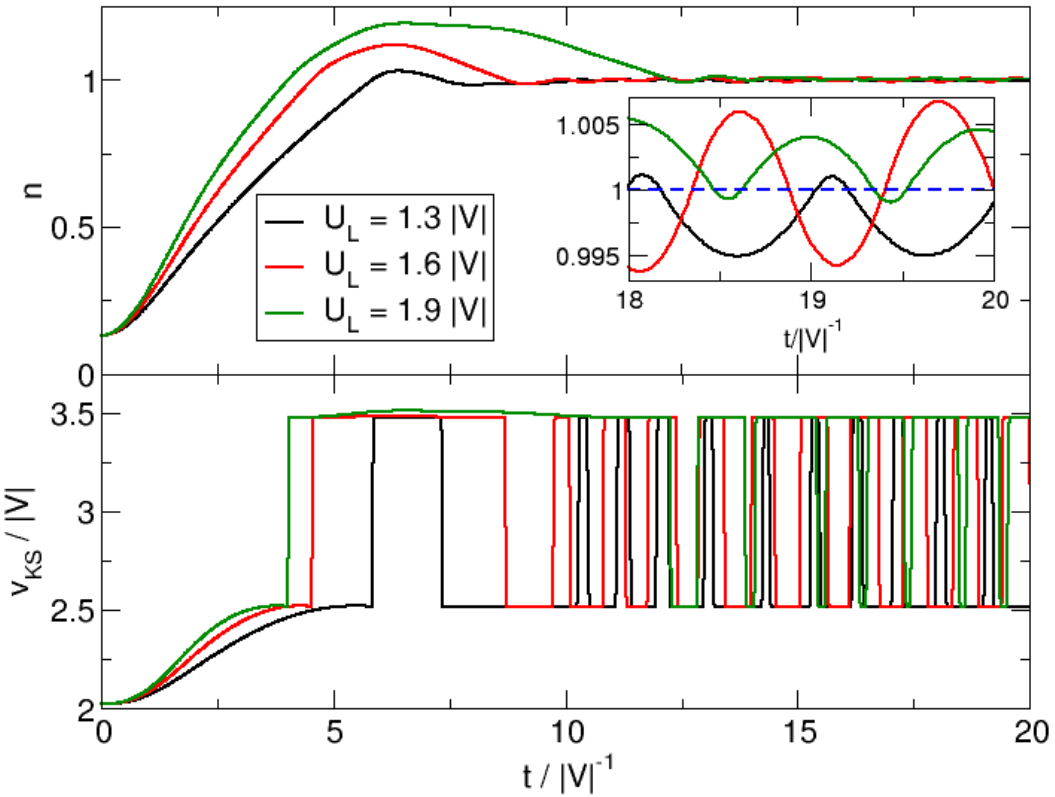
steady-state density as function of bias for different hoppings from lead to impurity



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)

TD density and KS potential in presence of discontinuity



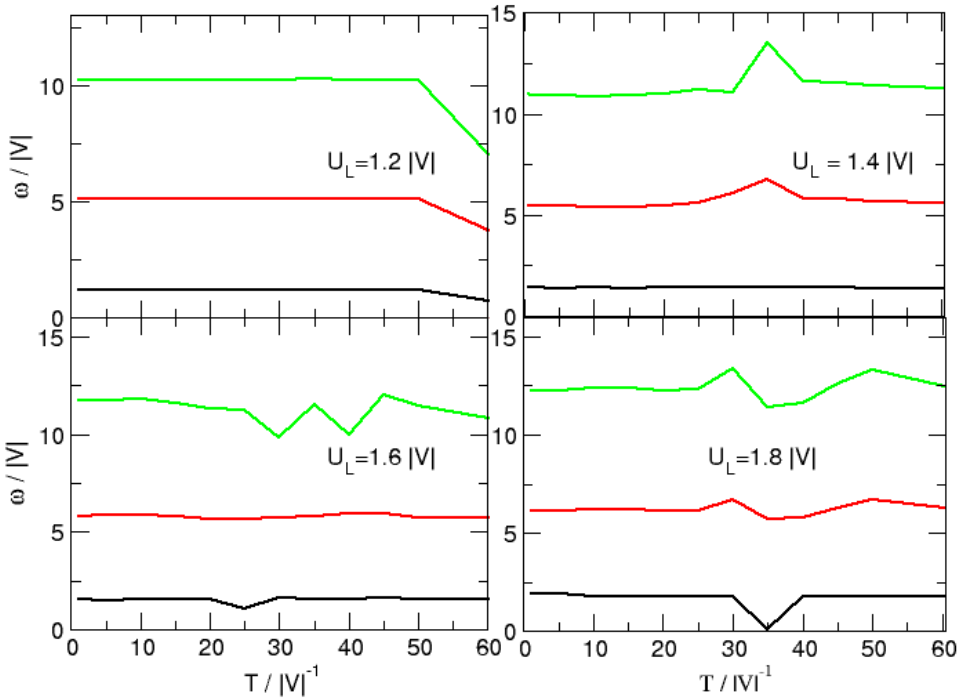
density shows small oscillations around integer occupation

TD KS potential: series of almost rectangular potential steps

History dependence of oscillations?

soft switching on of bias $U_L(t) = \begin{cases} U_0 \sin^2(\frac{\pi}{2T}t) & \text{for } t < T \\ U_0 & \text{otherwise} \end{cases}$

first main frequencies in Fourier transform of TD density for different switching times



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!

Collaborators:

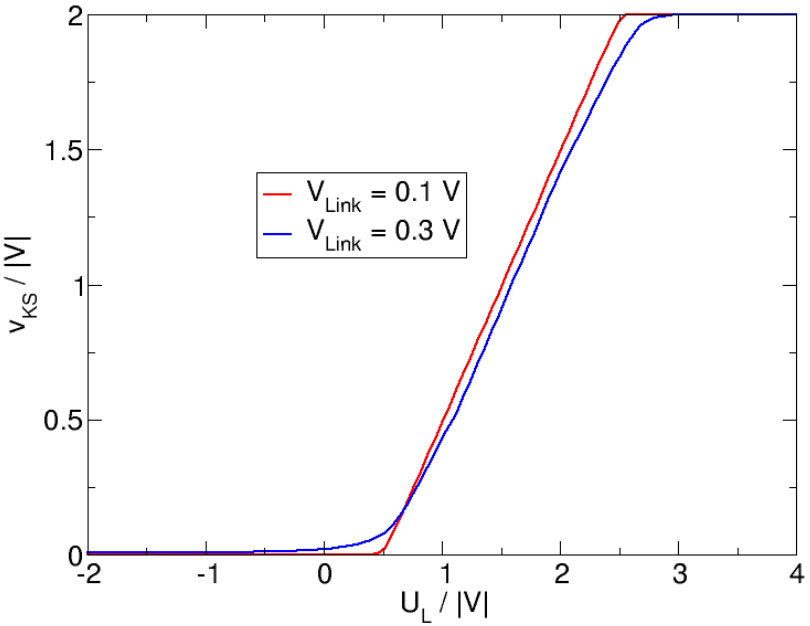
- G. Stefanucci, Univ. Tor Vergata, Rome, Italy
- A.-M. Uimonen and R. van Leeuwen, Univ. of Jyväskylä, Finland
- C. Verdozzi, Univ. Lund, Sweden
- E. Khosravi and E.K.U. Gross, MPI Halle, Germany

Summary

- TD approaches to transport: TDDFT and Kadanoff-Baym (MBPT, NEGF)
- Bistabilities: more than one steady state, TD switching between them
- Derivative discontinuity in TD transport: absence of steady state

KS potential of static approach

KS potential (static)



time-averaged KS potential for large times

$\lim_{t\to\infty} \frac{1}{T} \int_t^{t+T} \mathrm{d}t'\, v_{KS}(t')$

seems to agree with KS potential obtained from static approach