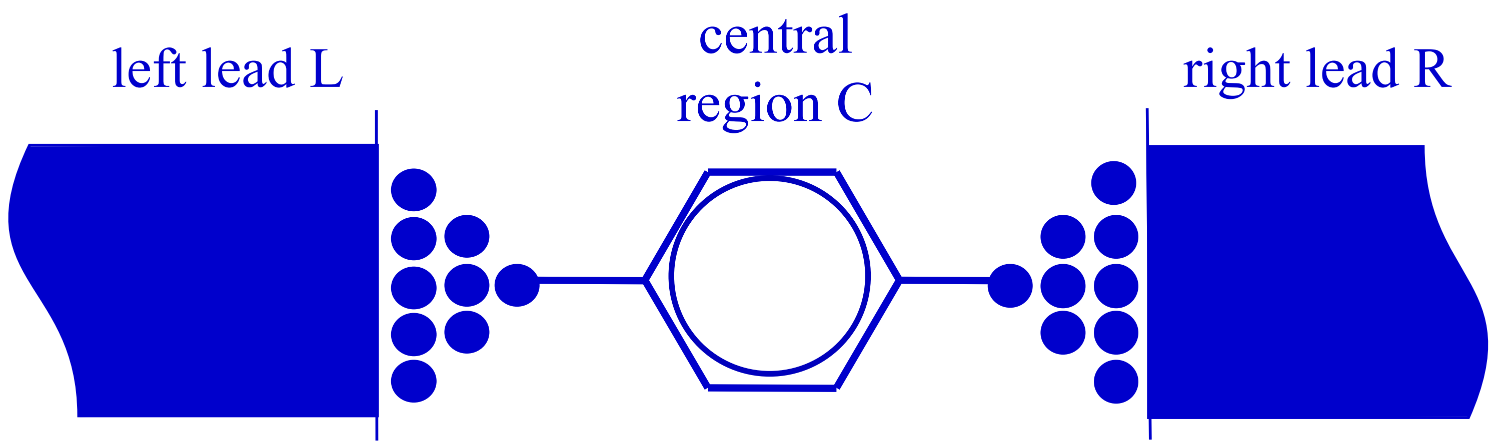
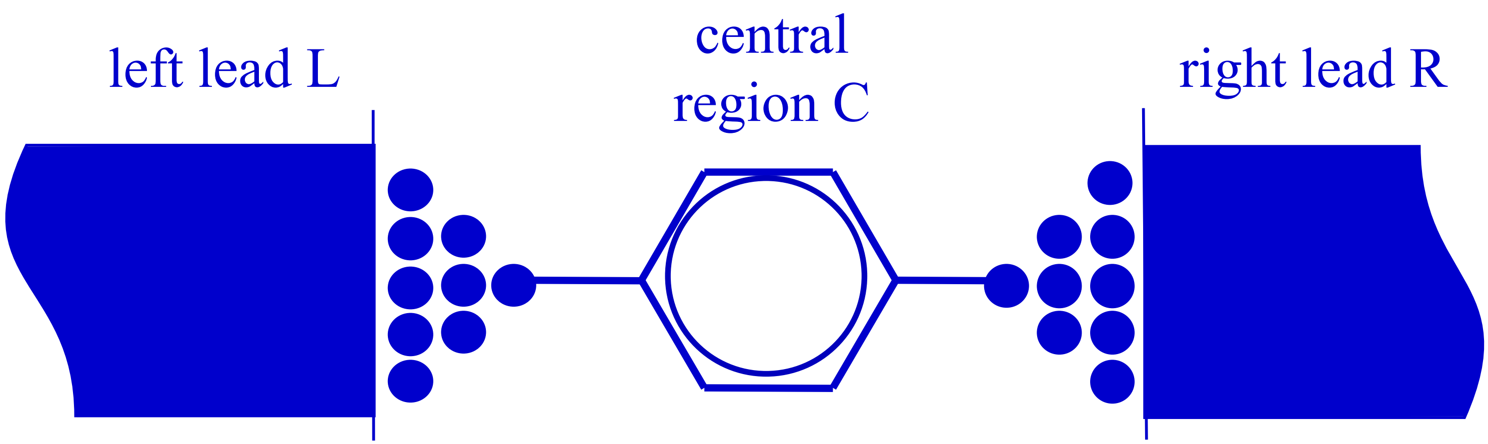
Beyond the linear regime: Real-time TDDFT prediction of electron dynamics far from equilibrium

Electronic transport with TDDFT



Bias between L and R is turned on: $U(t) \longrightarrow V$ for large t

Electronic transport with TDDFT

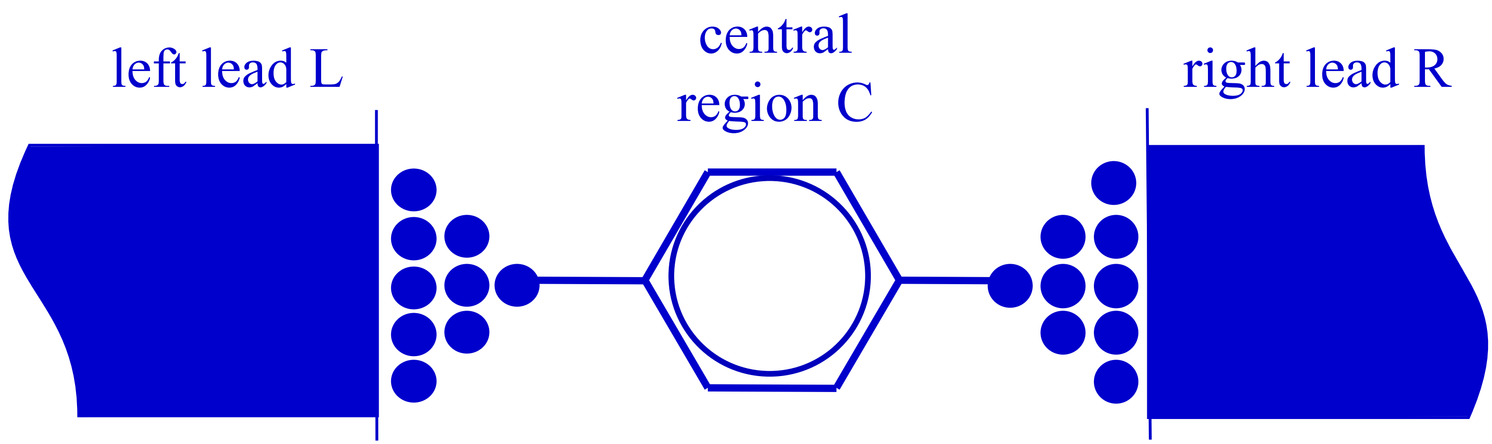


Bias between L and R is turned on: $U(t) \longrightarrow V$ for large t

Questions:

- After switching-on, does one always reach a steady state?
- Is the steady state unique?
- How to deal with time-dependent external fields?

Electronic transport with TDDFT

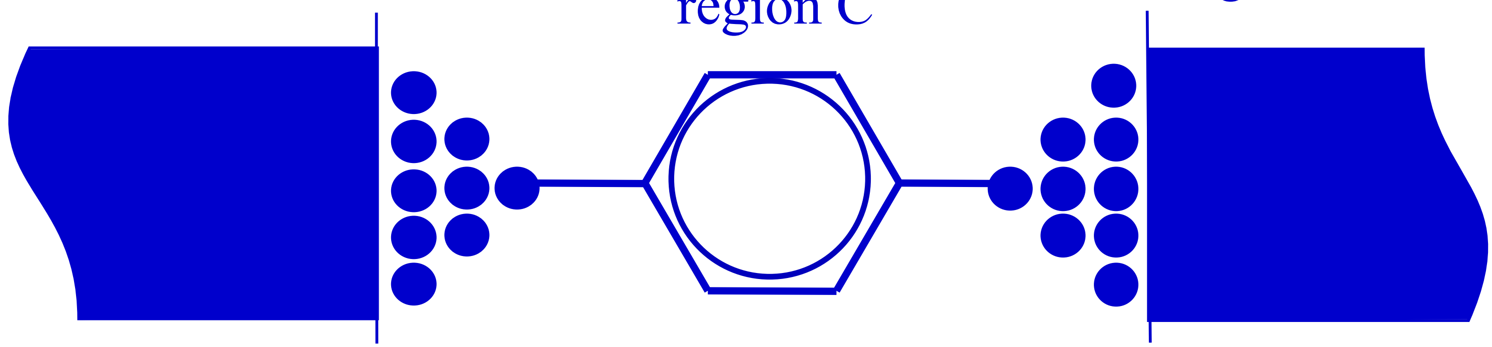


TDKS equation (E. Runge, EKUG, PRL **52**, 997 (1984))

$$i\hbar \frac{\partial}{\partial t} \varphi_j(rt) = \left(-\frac{\hbar^2 \nabla^2}{2m} + v_{KS}[\rho](rt) \right) \varphi_j(rt)$$

$$v_{KS}[\rho(r't')](rt) = v(rt) + \int d^3r' \frac{\rho(r't)}{|r - r'|} + v_{xc}[\rho\,(r't')](r\,t)$$

Electronic transport with TDDFT



TDKS equation

$$i\frac{\partial}{\partial t}\begin{pmatrix}\varphi_L(t)\\ \varphi_C(t)\\ \varphi_R(t)\end{pmatrix}=\begin{pmatrix}H_{LL}(t) & H_{LC}(t) & H_{LR}(t)\\ H_{CL}(t) & H_{CC}(t) & H_{CR}(t)\\ H_{RL}(t) & H_{RC}(t) & H_{RR}(t)\end{pmatrix}\begin{pmatrix}\varphi_L(t)\\ \varphi_C(t)\\ \varphi_R(t)\end{pmatrix}$$

Effective TDKS Equation for the central (molecular) region only

S. Kurth, G. Stefanucci, C.O. Almbladh, A. Rubio, E.K.U. Gross,
Phys. Rev. B 72, 035308 (2005)

$$
\begin{aligned}
i\frac{\partial}{\partial t}\varphi_C(t) = {} & H_{CC}(t)\varphi_C(t) \\
& + \int_0^t dt' \left[H_{CL} G_L(t,t') H_{LC} + H_{CR} G_R(t,t') H_{RC} \right] \varphi_C(t') \\
& + i H_{CL} G_L(t,0)\varphi_L(0) + i H_{CR} G_R(t,0)\varphi_R(0)
\end{aligned}
$$

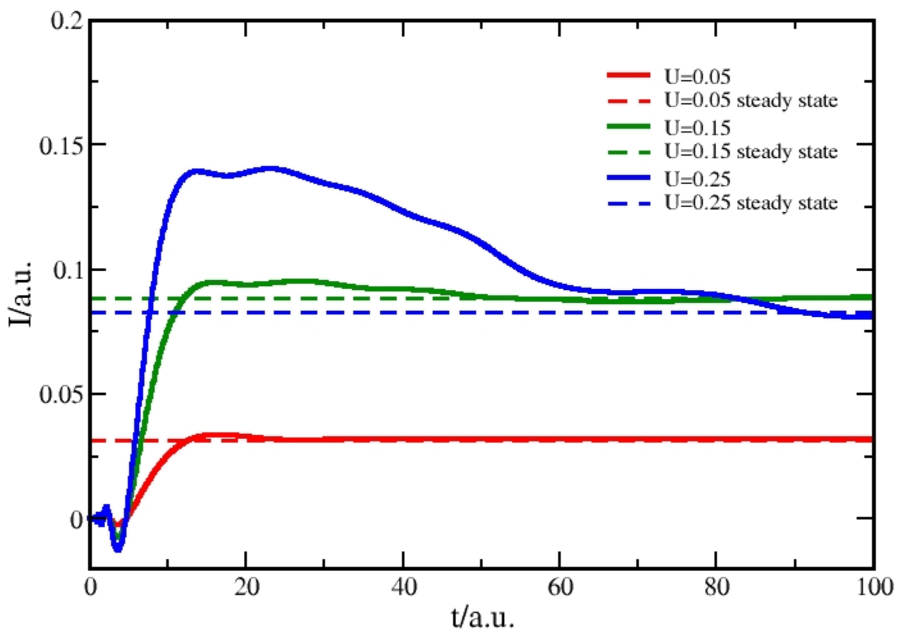
source term: $L \rightarrow C$ and $R \rightarrow C$ charge injection

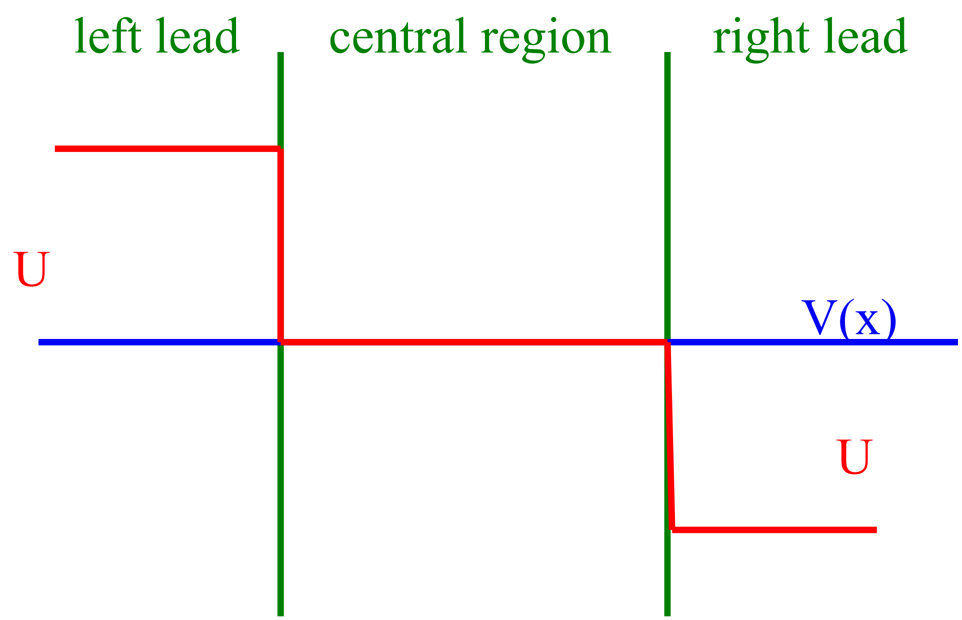
memory term: $C \rightarrow L \rightarrow C$ and $C \rightarrow R \rightarrow C$ hopping

Note: So far, no approximation has been made.

Numerical examples for non-interacting electrons

Recovering the Landauer steady state





Time evolution of current in response to bias switched on at time t = 0,

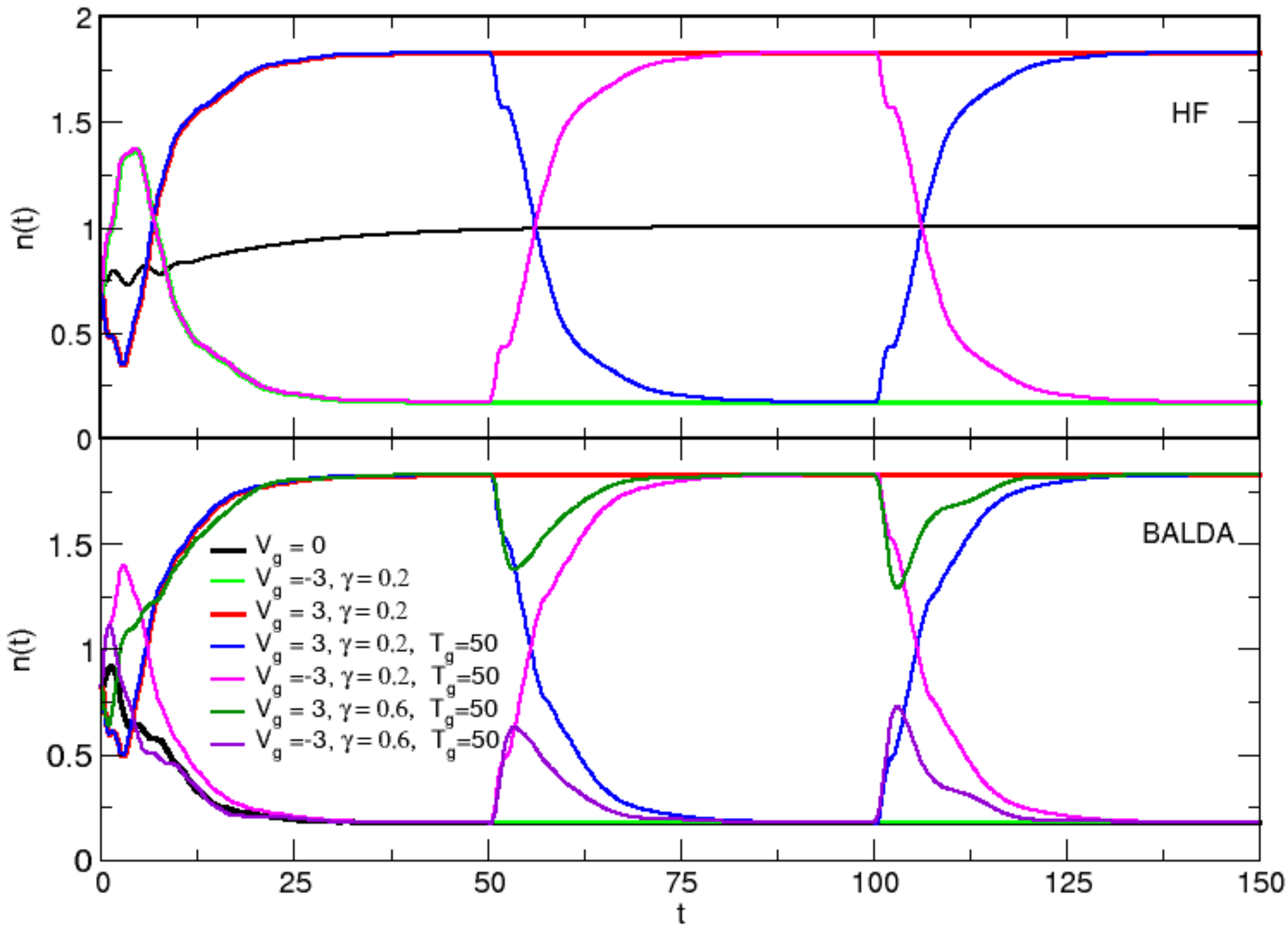
Fermi energy $\varepsilon_F = 0.3$ a.u.

Steady state coincides with Landauer formula

and is reached after a few femtoseconds

Can there be more than one steady state?

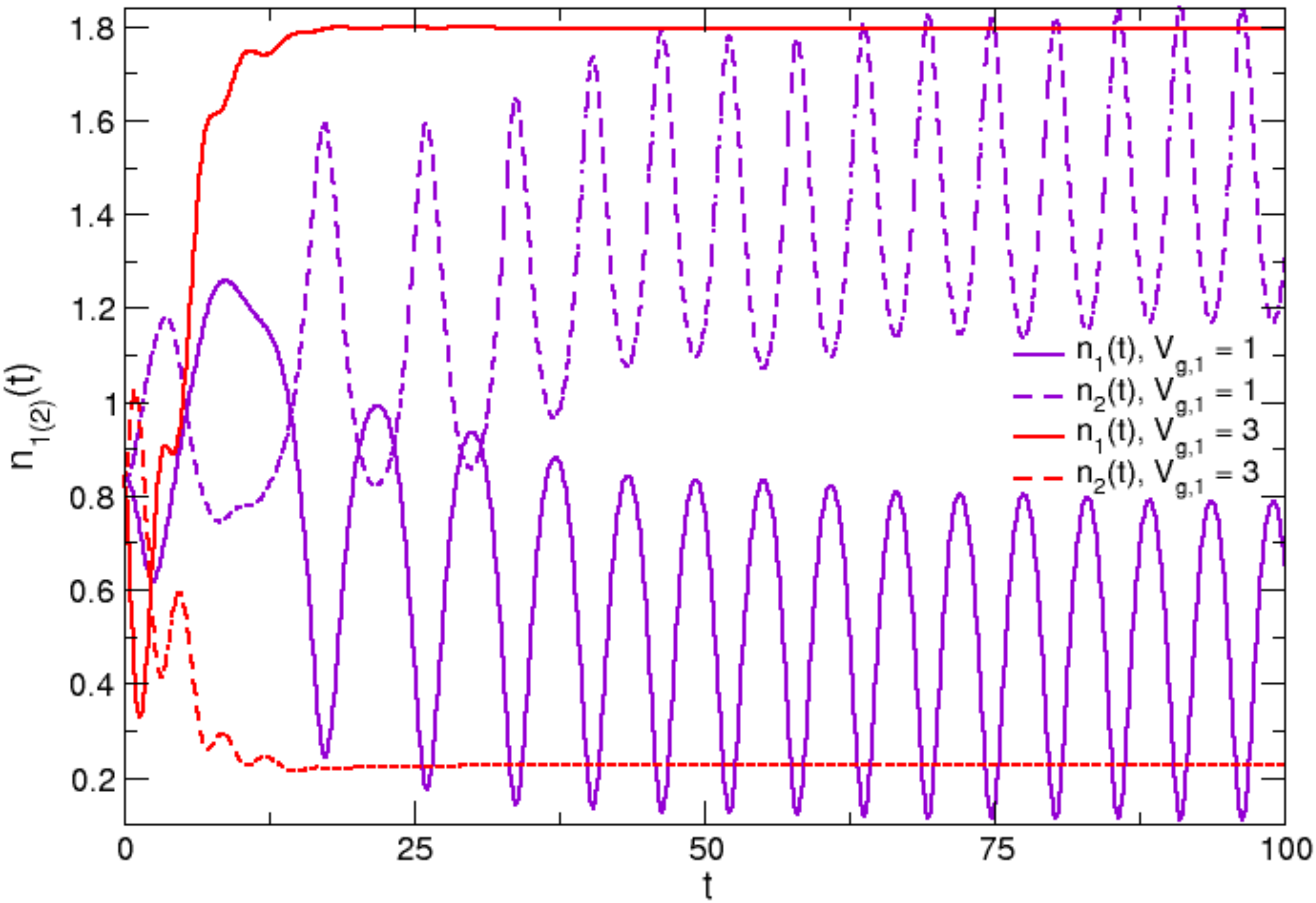
Multi-stability in TDHF and TDDFT for one-site Anderson model



E. Khosravi, A.M. Uimonen, A. Stan, G. Stefanucci, S. Kurth, R. van Leeuwen, E.K.U.G. Phys. Rev. B 85, 075103 (2012)

Is there always a steady state?

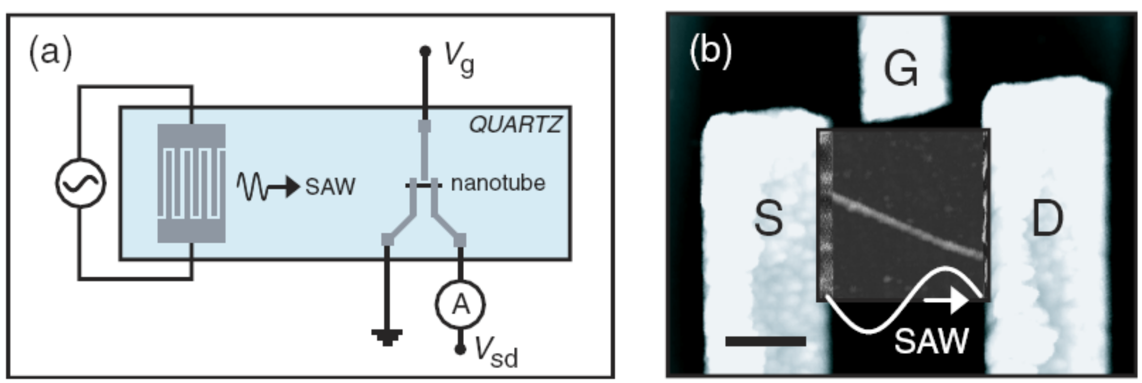
No steady state in two-site Anderson model



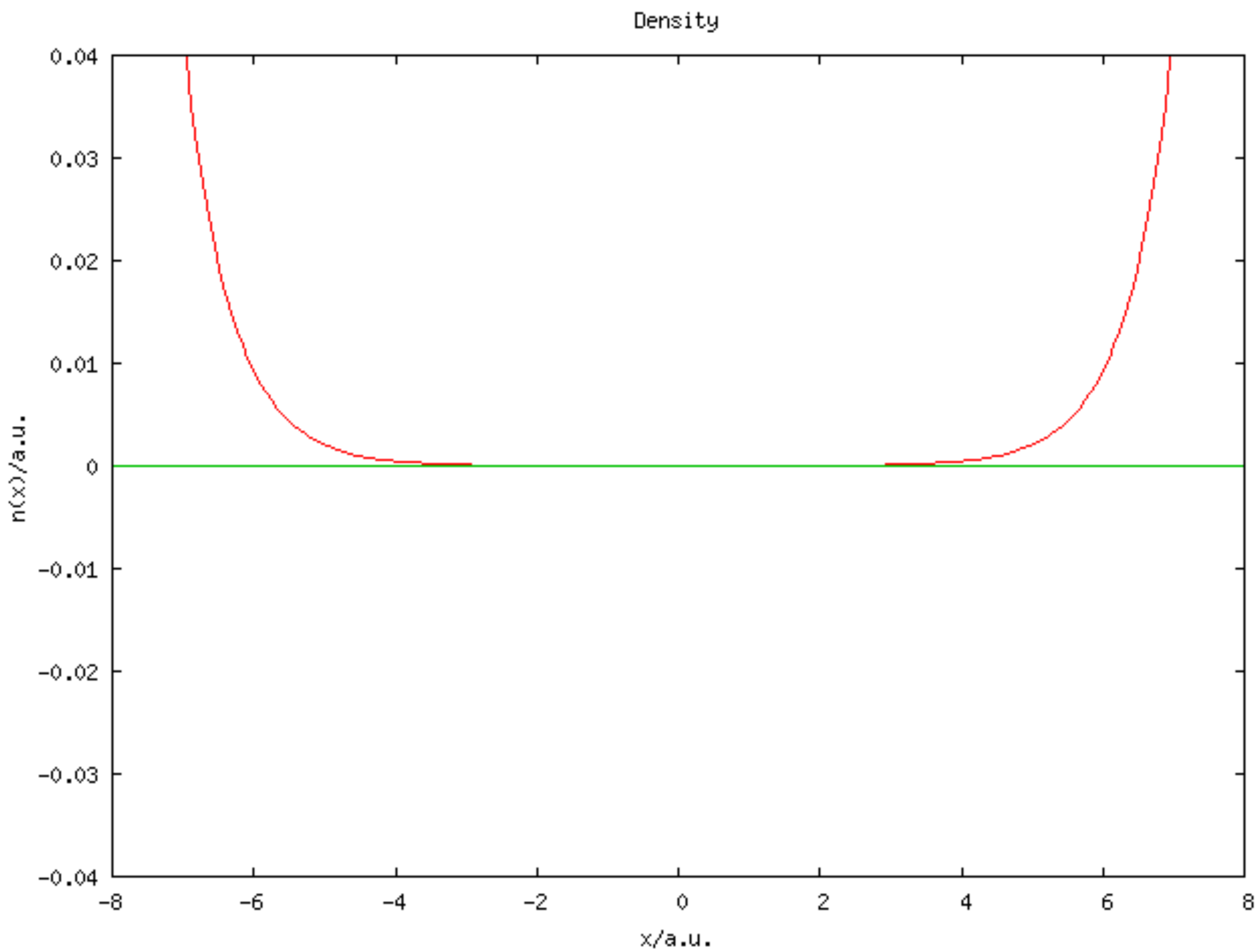
ELECTRON PUMP

Device which generates a net current between two electrodes (with no static bias) by applying a time-dependent potential in the device region

Experimental realization : Pumping through carbon nanotube by surface acoustic waves on piezoelectric surface (Leek et al, PRL 95, 256802 (2005))

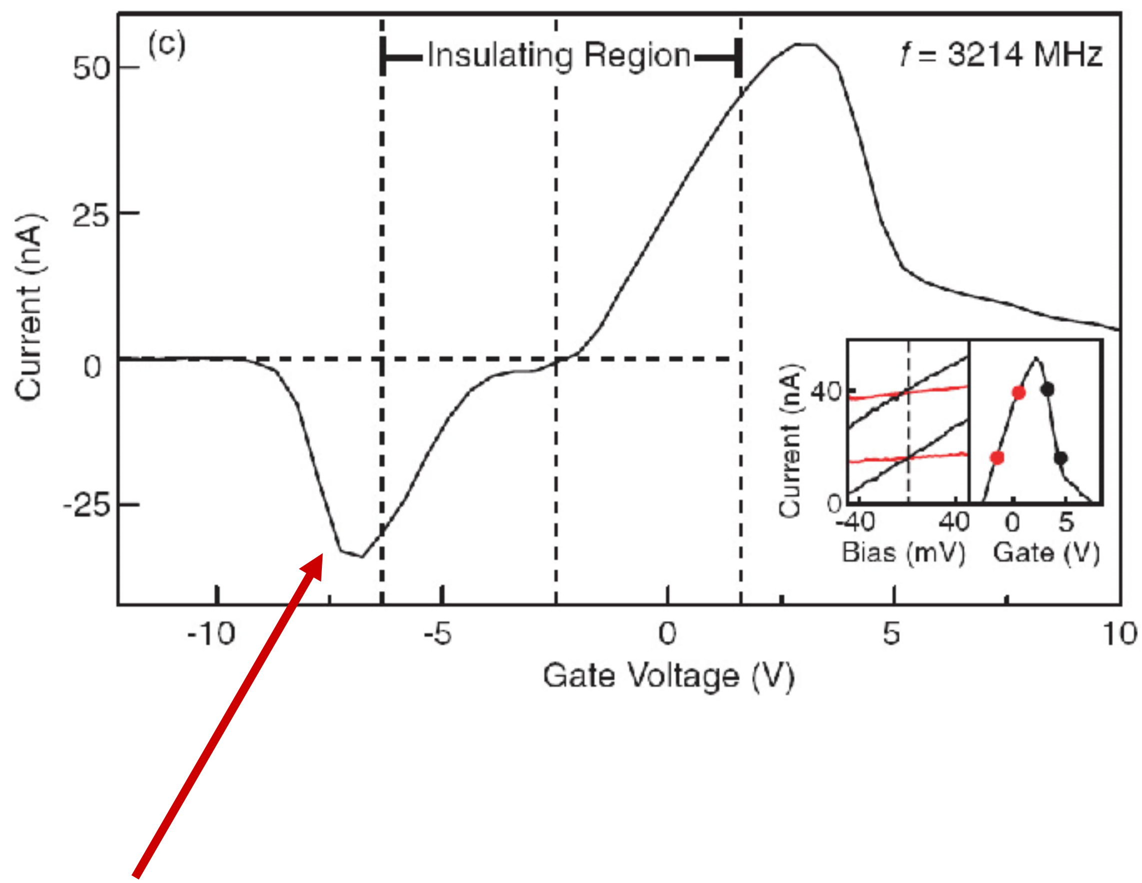


Pumping through a square barrier (of height 0.5 a.u.) using a travelling wave in device region
$U(x,t) = U_o \sin(kx-\omega t)$ ($k = 1.6$ a.u., $\omega = 0.2$ a.u. Fermi energy = 0.3 a.u.)

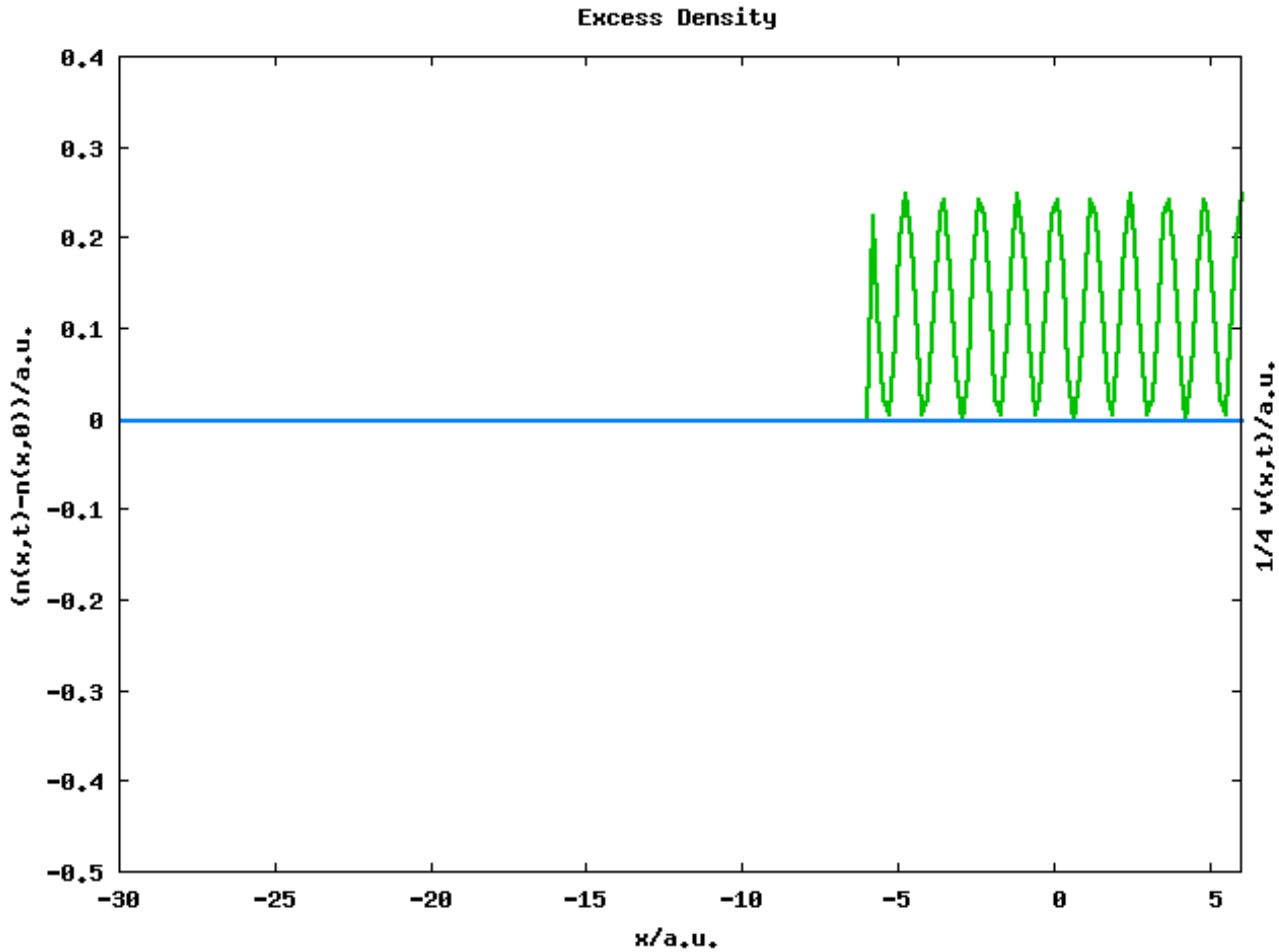


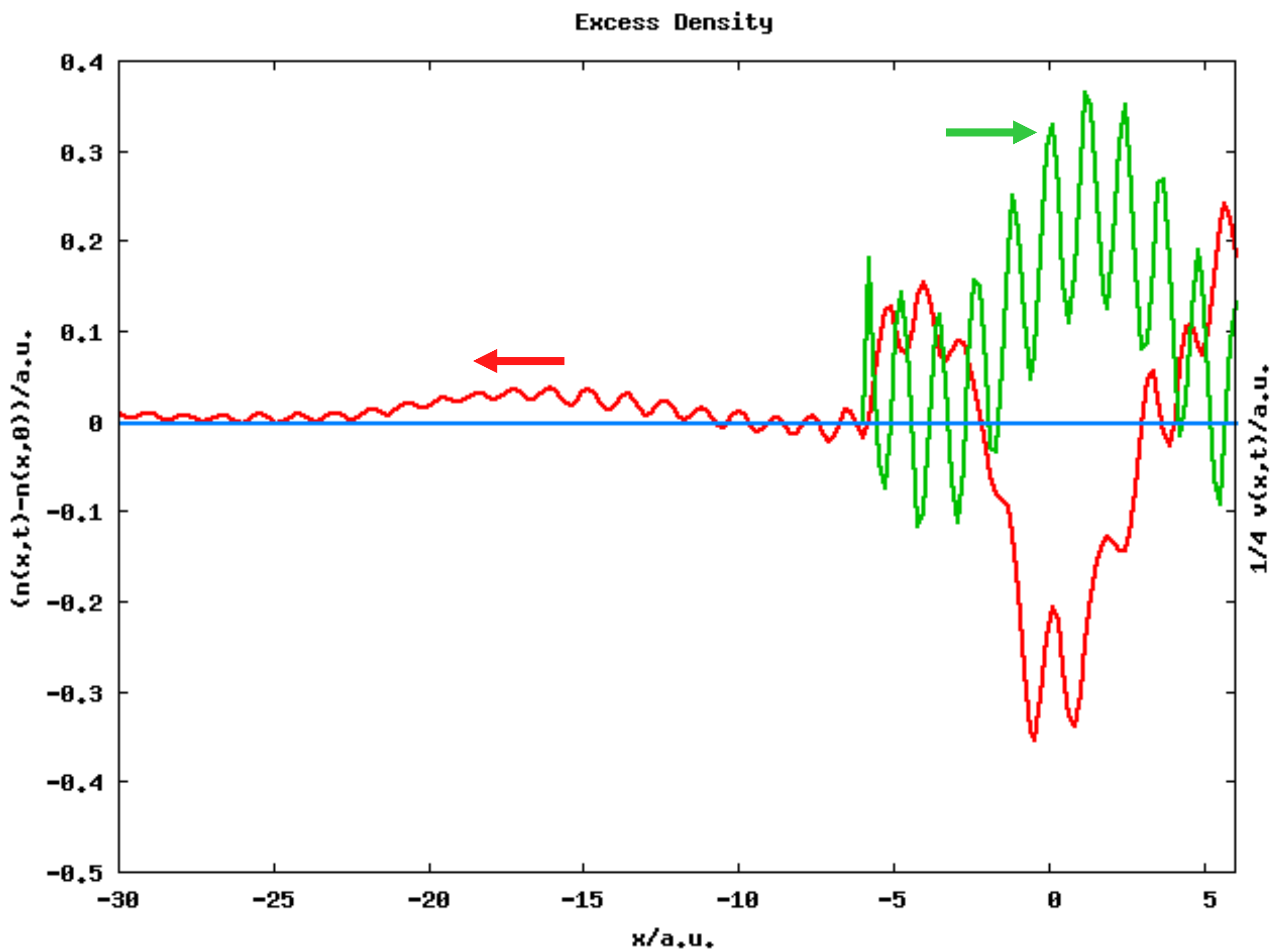
Archimedes' screw: patent 200 B.C.

Experimental result:



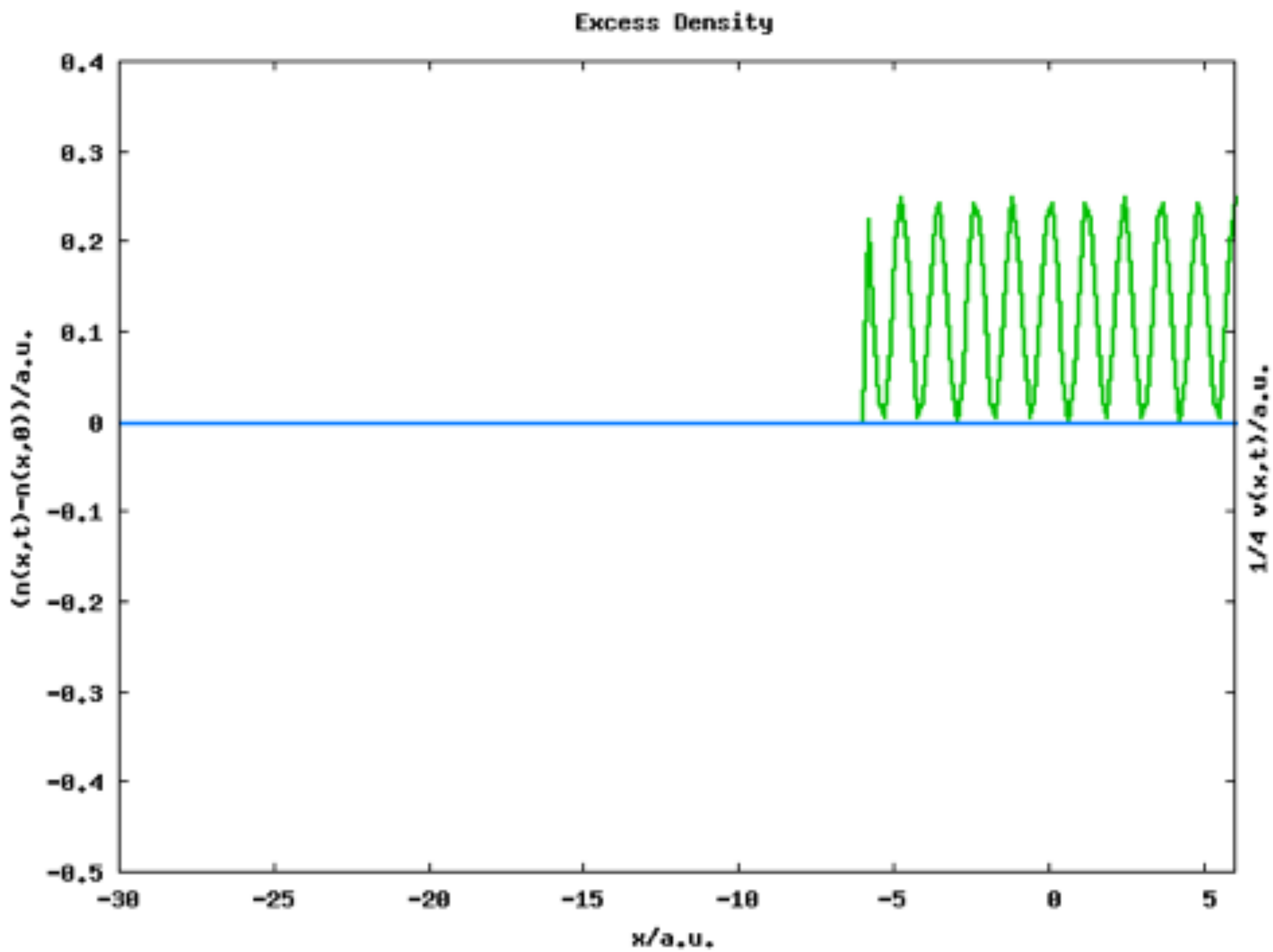
Current flows in direction opposite to sound wave


Excess Density
0.4
0.3
0.2
0.1
0
-0.1
-0.2
-0.3
-0.4
-0.5
-30
-25
-20
-15
-10
-5
0
5
x/a.u.
(n(x,t)-n(x,0))/a.u.
1/4 v(x,t)/a.u.



Current goes in direction opposite to the external field !!

G. Stefanucci, S. Kurth, A. Rubio, E.K.U. Gross, Phys. Rev. B 77, 075339 (2008)



G. Stefanucci, S. Kurth, A. Rubio, E.K.U. Gross, Phys. Rev. B 77, 075339 (2008)