Steady-state density functional theory for finite-bias conductances

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Coulomb Blockade in Transport at Finite Bias

Experimental conductances of graphene quantum dot as function of gate and bias (J. Güttinger et al, PRL 103, 046810 (2009))

Questions:
- Can we design a proper DFT framework to calculate finite-bias conductances?
- If yes, what are the essential features for the xc potential(s) to describe the Coulomb blockade diamonds?
Outline

- Steady-state DFT formalism (i-DFT) for electron transport
  - Choice of variables and 1-1 map
  - Kohn-Sham system: Hxc gate and xc bias potentials
- Finite-bias Coulomb blockade: requirements on xc potentials
  - Single impurity Anderson model
  - Constant-Interaction Model for multiple levels
  - Approximate xc potentials for general case
- Finite-bias Coulomb blockade for benzene
- Summary
i-DFT for steady-state electron transport

Schematic transport setup with arbitrary molecular region $\mathcal{R}$ (with molecular potential $v(r)$) and applied bias $V$, interested only in \textbf{steady state}

Choice of variables for steady-state DFT:

molecular steady-state density $n(r)$ in region $\mathcal{R}$ and steady-state current $I$ through $\mathcal{R}$
**Theorem:**
For any finite temperature the map \((v(r), V) \rightarrow (n(r), I))\) is invertible in a finite, gate-dependent window around bias \(V = 0\).

**Proof:** show that Jacobian

\[
J_0 = \text{Det} \begin{pmatrix}
\frac{\delta n(r)}{\delta v(r')} & \frac{\partial n(r)}{\partial V} \\
\frac{\delta I}{\delta v(r')} & \frac{\partial I}{\partial V}
\end{pmatrix}_{V=0}
\]

is non-vanishing.

**note:**
\[
\frac{\delta I}{\delta v(r')} \bigg|_{V=0} = 0 \quad \text{since at zero bias a variation of the gate does not induce a steady current.}\
\]
Lehmann representation of static density response function

$$\chi_0(r, r') = \frac{\delta n(r)}{\delta v(r')} \bigg|_{V=0} \text{ at finite temperature}$$

$$\chi_0(r, r') = \frac{1}{Z} \sum_{i,j} \frac{f_{ij}(r)f_{ij}(r')}{\Omega_{ij}^2 + \eta^2} \Omega_{ij} \left( e^{-\beta E_i} - e^{-\beta E_j} \right) e^{\beta \mu N_i}$$

with partition function $Z$, $f_{ij}(r) = \langle \Psi_i | \hat{n}(r) | \Psi_j \rangle - \delta_{ij} n(r)$, and $\Omega_{ij} = E_i - E_j \rightarrow$ for $E_i \leq E_j$, $\Omega_{ij} \leq 0$ and $(e^{-\beta E_i} - e^{-\beta E_j}) \geq 0 \quad \rightarrow$ Det[$\chi_0$] < 0 for all $v(r)$. 

Similarly, using Lehmann representation of $G_0 = \frac{\partial I}{\partial V} \bigg|_{V=0}$ (D. Bohr et al, EPL 73, 246 (2006)) one finds that $G_0 > 0 \quad \rightarrow J_0 = \text{Det}[\chi_0] G_0 < 0$ for all gates $v(r)$. 
let \((n, I)\) be the density and steady-state current of an interacting system with gate potential and bias \((v, V)\). Under assumption of non-interacting \(v\)-representability there is a unique pair of potentials \((v_s, V_s)\) which reproduces \((n, I)\) in a non-interacting system →

**Hxc gate and xc bias potentials**

\[
\begin{align*}
v_{Hxc}[n, I](r) &= v_s[n, I](r) - v[n, I](r) \\
V_{xc}[n, I] &= V_s[n, I] - V[n, I]
\end{align*}
\]
i-DFT self-consistent KS equations

\[ n(r) = 2 \sum_{\alpha=L,R} \int \frac{d\omega}{2\pi} f\left(\omega + s_\alpha \frac{V + V_{xc}}{2}\right) A_\alpha(r,\omega) \]

\[ I = 2 \sum_{\alpha=L,R} \int \frac{d\omega}{2\pi} f\left(\omega + s_\alpha \frac{V + V_{xc}}{2}\right) s_\alpha T(\omega) \]

with KS partial spectral function
\[ A_\alpha(r,\omega) = \langle r|G_s(\omega)\Gamma_\alpha(\omega)G_s^\dagger(\omega)|r\rangle, \] KS transmission function
\[ T(\omega) \] and \[ s_{R/L} = \pm 1 \]

Note: equivalent to Landauer+DFT formalism if \( V_{xc} \) set to zero
i-DFT expression for zero-bias conductance

linearize i-DFT selfconsistency conditions $\rightarrow$

exact expression for zero-bias conductance

$$G_0 = \frac{G_{0,s}}{1 - G_{0,s} \frac{\partial V_{xc}}{\partial I} \bigg|_{I=0}}$$

with zero-bias KS conductance $G_{0,s}$.

Remark: The $G_{0,s}$ of i-DFT is exactly the same as the usual Landauer-DFT zero-bias KS conductance!
construct $xc$ potentials for single impurity Anderson model: single site (energy $v$) with a charging energy $U$ for double occupancy coupled (coupling strength $\gamma$) to two leads (wide band limit, WBL)
Single impurity Anderson model (SIAM)

Density and current from spectral function (in WBL)

\[
N = \int \frac{d\omega}{2\pi} \left[ f_\beta(\omega - V/2) + f_\beta(\omega + V/2) \right] A(\omega)
\]

\[
I = \frac{\gamma}{2} \int \frac{d\omega}{2\pi} \left[ f_\beta(\omega - V/2) - f_\beta(\omega + V/2) \right] A(\omega)
\]

Model (interacting) spectral function of site connected to leads

\[
A(\omega) = \frac{N}{2} L_\gamma(\omega - \nu - U) + \left(1 - \frac{N}{2}\right) L_\gamma(\omega - \nu)
\]

with Lorentzian \(L_\gamma(\omega) = \frac{\gamma}{\omega^2 + (\frac{\gamma}{2})^2}\)

Reasonable in Coulomb blockade regime, i.e., for temperatures above the Kondo temperature \(T_K\)
SIAM: reverse engineering for xc potentials

Remark:
model spectral function gives exactly the same $N$ and $I$ as the rate equation approach (C.W.J. Beenakker, PRB 44, 1646 (1991))

Reverse engineering for xc potentials:
for given $(N, I)$: numerically find the potentials $(v, V)$ which yield this $(N, I)$ first for the interacting case, then find non-interacting potentials $(v_s, V_s)$ which also yield same densities $\rightarrow$ construct $(H)_{xc}$ potentials as

$$\left(v_{Hxc}[N, I], V_{xc}[N, I]\right) = \left(v_s[N, I] - v[N, I], V_s[N, I] - V[N, I]\right)$$
SIAM: Hxc gate and xc bias potentials

SIAM (H)xc potentials from reverse engineering

Domain: $|I| \leq \frac{\gamma}{2}N$ for $N \in [0, 1]$ and $|I| \leq \frac{\gamma}{2}(2 - N)$ for $N \in [1, 2]$
SIAM: Hxc gate and xc bias potentials

- Hxc gate (xc bias) has smeared steps of height $U/2$ ($U$)
- DFT xc discontinuity at $N = 1$ in $v_{Hxc}[N, I = 0]$ bifurcates as current starts flowing
- xc bias has opposite sign of current, i.e., xc bias counteracts external bias
SIAM: Hxc gate and xc bias potentials

simple parametrization (low temperature regime)

**parametrization of xc potentials for SIAM**

\[
\nu\text{Hxc}[N, I] = \frac{U}{4} \sum_{s=\pm} \left[ 1 + \frac{2}{\pi} \arctan \left( \frac{N + sI - 1}{W} \right) \right]
\]

\[
V\text{xc}[N, I] = -U \sum_{s=\pm} \frac{s}{\pi} \arctan \left( \frac{N + sI - 1}{W} \right)
\]

parameter \(W = 0.16\gamma/U\) to obtain best fit to reverse-engineered xc potentials
Consider CIM with $M$ degenerate single-particle (sp) levels ($\text{SIAM} \rightarrow M = 1$); same coupling $\gamma$ to leads of all sp levels.

All sp levels equivalent $\rightarrow$ $H_{xc}$ gate and $xc$ bias depend only on total number of electrons $N$ and $I$. 
Hxc potentials by reverse engineering of \((N, I)\) as function of \((v, V)\) obtained from Beenakker's rate equations (RE)

- again smeared steps of height \(U/2\) \((U)\) for Hxc gate (xc bias)
- edges at piecewise linear functions of \(\Delta_K^{(\pm)}(N, I)\)
- vertices in \((N, I)\) plane correspond to plateau values of \((N, I)\) in gate-bias scan of RE; can be obtained analytically
parametrization of degenerate CIM xc potentials

simple parametrization (low temperature regime)

\[ v_{Hxc}^{(M)}[N, I] = \frac{U}{4} \sum_{K=1}^{2M-1} \sum_{s=\pm} \left[ 1 + \frac{2}{\pi} \tan^{-1} \left( \frac{\Delta_K^{(s)}(N, I)}{W} \right) \right] \]

\[ V_{xc}^{(M)}[N, I] = -U \sum_{K=1}^{2M-1} \sum_{s=\pm} \frac{s}{\pi} \tan^{-1} \left( \frac{\Delta_K^{(s)}(N, I)}{W} \right) \]

\( \Delta_K^{(\pm)}(N, I) \) piecewise linear function of \( N \) and \( I \) which vanishes at step edges passing through \( (N = K, I = 0) \) with positive (\( s = + \)) and negative (\( s = - \)) slopes
parametrization of CIM xc potentials for arbitrary sp level structure

\[ v_{Hxc}[n,I] = \sum_{p=1}^{D} v^{(M_p)}_{Hxc} [N - N_p, I] + \frac{U}{4} \sum_{p=1}^{D-1} \sum_{s=\pm} \left[ 1 + \frac{2}{\pi} \tan^{-1} \left( \frac{N + \frac{2s}{\gamma} I - N_{p+1}}{W} \right) \right] \]

and similarly for xc bias \[ V_{xc}[n,I] \]

\( D \): number of different sp energies (not counting degeneracies)
\( M_p \): degeneracy of \( p \)-th sp level
\( N_p \): maximal number of particles in first \( p - 1 \) degenerate levels
model benzene as a 6-level CIM with $\varepsilon_1 = -\varepsilon_6 = -5.08$ eV and $\varepsilon_2 = \varepsilon_3 = -\varepsilon_4 = -\varepsilon_5 = -2.54$ eV; $U = 0.5$ eV
solve i-DFT equations for $(N, I)$ with model xc potentials, calculate differential conductance map $\frac{dI}{dV}$ as function of $(v, V)$
LB-DFT, i.e. neglecting xc bias, gives spurious Kondo plateaus at finite bias; low-bias CB not correctly described
i-DFT in simple approx. gets all low-bias features of the RE
size of RE matrix scales as $4^{N_{lev}}$; simple i-DFT doesn’t scale at all (2 coupled nonlinear eqs. for $N$ and $I$)
Summary

i-DFT: DFT formulation for steady-state transport at arbitrary bias

- i-DFT based on 1-1 map (for small bias) between \((n(\mathbf{r}), I)\) and \((v(\mathbf{r}), V)\)
- i-DFT KS formulation with both Hxc gate and xc bias
- xc potentials from models to describe Coulomb blockade: step structures in xc potentials; xc derivative discontinuity of equilibrium DFT bifurcates as current starts flowing
- simple model xc potentials with correct step structure are sufficient to describe finite-bias CB correctly for not too large bias

Reference:

- Stefanucci, Kurth, arXiv:1505.07354