Steady-State Density Functional Theory for Many-Body Spectral Functions

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Outline

- Many-body spectral functions from electron transport
- Steady-state DFT formalism (i-DFT) for electron transport
- i-DFT xc potentials and spectral functions for model systems
- Mott metal-insulator transition from i-DFT

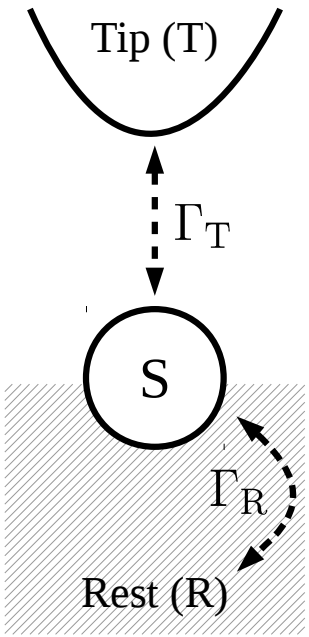
Many-body Spectral Function from electron transport

Reference:

- D. Jacob, S. Kurth, Nano Lett. **18**, 2086 (2018)

Ideal STM setup to extract spectral function from I-V

STM-like setup



Interacting system S (weakly) coupled to STM tip (T) via Γ_T and coupled via Γ_R to the rest (R) of the system.

"Ideal STM setup": bias voltage V drops entirely at the tip and take the limit of infinitesimaly weak coupling to the tip, i.e., $\Gamma_T \to 0$

Meir-Wingreen formula for steady current

Meir-Wingreen formula for steady current from tip T to interacting system S (PRL **68**, 2512 (1992))

Meir-Wingreen formula for steady current

$$I(V) = 2\int \frac{\mathrm{d}\omega}{2\pi} \mathrm{Tr}\left\{ f(\omega - V)\mathbf{\Gamma}_{\mathrm{T}}\mathbf{A}(\omega) + i\mathbf{\Gamma}_{\mathrm{T}}\mathbf{G}^{<}(\omega)\right\}$$

with Fermi function $f(\omega)$, (non-equilibrium) many-body spectral function $\mathbf{A}(\omega)$ and lesser Green function $\mathbf{G}^{<}(\omega)$
$\longrightarrow$ differential conductance in zero-temperature limit

$$\frac{\partial I}{\partial V} \xrightarrow[\mathbf{\Gamma}_{\mathrm{T}}\to 0]{} \int \frac{d\omega}{\pi} \frac{\partial f_{\mathrm{T}}(\omega - V)}{\partial V} \mathrm{Tr}\left[\mathbf{\Gamma}_{\mathrm{T}}\mathbf{A}(\omega)\right] \xrightarrow[T\to 0]{} \frac{\mathrm{Tr}\left[\mathbf{\Gamma}_{\mathrm{T}}\mathbf{A}(V)\right]}{\pi}$$

Spectral function from differential conductance

for coupling matrix $\mathbf{\Gamma}_{\mathrm{T}} = \gamma_m^{\mathrm{T}} |m\rangle\langle m|$

Equilibrium spectral function from differential conductance

$$A_m(\omega) = \lim_{\gamma_m^{\mathrm{T}} \to 0} \frac{\pi}{\gamma_m^{\mathrm{T}}} \frac{\partial I}{\partial V}\bigg|_{V=\omega}$$

note:
in ideal STM limit of vanishing coupling to the tip the system is essentially unperturbed by the bias and the spectral function becomes the *equilibrium* spectral function of the interacting system S in contact with the rest R

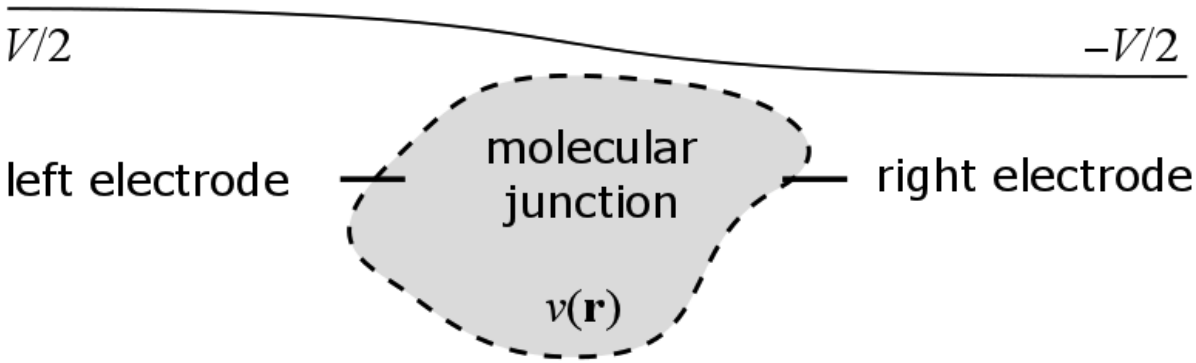
i-DFT for steady-state electron transport

References:

- G. Stefanucci, S. Kurth, Nano Lett. **15**, 8020 (2015)
- S. Kurth, G. Stefanucci, J. Phys.: Condens. Matter **29**, 413002 (2017)
- D. Jacob, S. Kurth, Nano Lett. **18**, 2086 (2018)

i-DFT for steady-state electron transport

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



Choice of variables for steady-state DFT:

molecular steady-state density $n(\mathbf{r})$ in region $\mathcal{R}$ and steady-state current I through $\mathcal{R}$

i-DFT for steady-state electron transport

Theorem:
For any *finite* temperature the map $(v(\mathbf{r}), V) \to (n(\mathbf{r}), I))$ is invertible in a finite, gate-dependent window around bias $V = 0$.

Density Functionalization: Find non-interacting system with potentials (v_s, V_s) which reproduces "densities" (n, I) of *interacting* system with potentials (v, V)
$\longrightarrow$ need for two (H)xc potentials

Hxc gate and xc bias potentials

$$v_{\mathrm{Hxc}}[n, I](\mathbf{r}) = v_s[n, I](\mathbf{r}) - v[n, I](\mathbf{r})$$

$$V_{\mathrm{xc}}[n, I] = V_s[n, I] - V[n, I]$$

i-DFT self-consistent KS equations

i-DFT KS equations for density and steady current

$$n(\mathbf{r}) = 2\sum_{\alpha=L,R}\int\frac{\mathrm{d}\omega}{2\pi}f_\beta\left(\omega + s_\alpha\frac{V+V_{\mathrm{xc}}}{2}\right)A_{\alpha,s}(\mathbf{r},\omega)$$

$$I = 2\sum_{\alpha=L,R}\int\frac{\mathrm{d}\omega}{2\pi}f_\beta\left(\omega + s_\alpha\frac{V+V_{\mathrm{xc}}}{2}\right)s_\alpha T_s(\omega)$$

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

Note: equivalent to Landauer+DFT formalism if V_{xc} set to zero and v_{Hxc} assumed to be independent of current

i-DFT equations in ideal STM limit

i-DFT KS equations for asymmetrically applied bias in STM limit $\Gamma_{\mathrm{T}} \longrightarrow 0$

i-DFT KS equations in ideal STM limit

$$n(\mathbf{r}) = 2\int \frac{\mathrm{d}\omega}{2\pi} f_\beta(\omega) A_{R,s}(\mathbf{r},\omega)$$

$$I = 2\int \frac{\mathrm{d}\omega}{2\pi} \left[f_\beta(\omega - V_s) - f_\beta(\omega)\right] \mathrm{Tr}\left\{\Gamma_{\mathrm{T}} A_{R,s}(\omega)\right\}$$

with KS Green function $G_s = \left(\omega - \frac{V_{\mathrm{xc}}}{2} - \mathbf{h}_s - \mathbf{\Sigma}_R(\omega)\right)^{-1}$ entering $A_{R,s}$ and *equilibrium* KS Hamiltonian $\mathbf{h}_s$

note: in STM limit, the i-DFT self-consistency conditions for n and I decouple completely

Many-body spectral function in terms of KS one

for choice of coupling matrix to tip $\Gamma_{\rm T} = \gamma_m^{\rm T} |m\rangle\langle m|$, take limit $\gamma_m^{\rm T} \to 0$ to relate KS to many-body spectral function

relation between KS and many-body spectral functions

$$A_m(\omega) = \lim_{\gamma_m^{\rm T}\to 0} \frac{A_{m,s}(\omega + V_{\rm xc})}{1 - \frac{\gamma_m^T}{\pi} \frac{\partial V_{\rm xc}}{\partial I} A_{m,s}(\omega + V_{\rm xc})}$$

where the current I entering $V_{\rm xc}$ is to be computed at bias $V = \omega$ and A_m and $A_{m,s}$ are local equilibrium many-body and KS spectral functions of the system S coupled to the rest R

i-DFT xc potentials and spectral functions for model systems

References:

- G. Stefanucci, S. Kurth, Nano Lett. **15**, 8020 (2015)
- S. Kurth, G. Stefanucci, Phys. Rev. B. **94**, 241103 (2016)(R)
- D. Jacob, S. Kurth, Nano Lett. **18**, 2086 (2018)
- S. Kurth, D. Jacob, Eur. Phys. J. B **91**, 101 (2018)

Model systems: single impurity and double dot

consider two models connected to wide-band lead (coupling γ)

Single impurity Anderson model (SIAM)

$$\hat{H}_{\mathrm{S}} = v\hat{n} + U\hat{n}_{\uparrow}\hat{n}_{\downarrow}$$

with $\hat{n} = \hat{n}_{\uparrow} + \hat{n}_{\downarrow}$

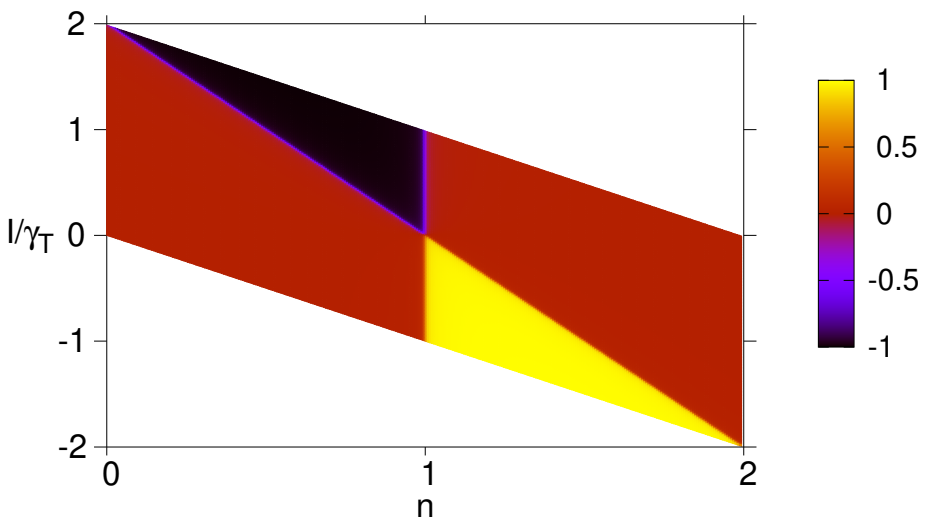
Double dot with density-density interaction

$$\hat{H}_{\mathrm{S}} = \sum_{j=1}^{2} v_j \hat{n}_j + \sum_{j=1}^{2} U_j \hat{n}_{j\uparrow}\hat{n}_{j\downarrow} + U_{12}\hat{n}_1\hat{n}_2$$

special case: $U_1 = U_2 = U_{12} = U$ Constant Interaction Model (CIM)

SIAM xc bias in Coulomb blockade regime

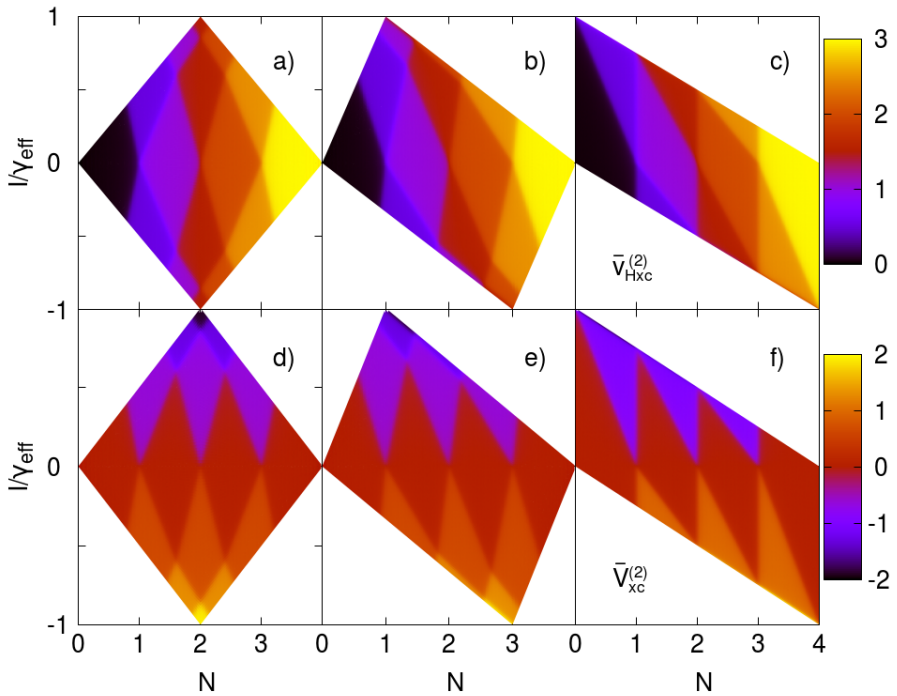
obtain xc bias by reverse engineering of Beenakker's rate equations (RE) (PRB **44**, 1646 (1991)) (valid in Coulomb blockade regime)



- parameters: $U = 1$, $\gamma = 0.02$
- xc bias has smeared steps of height U
- xc bias has opposite sign of current, i.e., xc bias counteracts external bias

CIM with M=2: Hxc gate and xc bias potentials

CIM xc potentials by reverse engineering of RE; from symmetric to asymmetric coupling ($\gamma_{\rm eff} = 4\gamma_L\gamma_R/\gamma$ where $\gamma = \gamma_L + \gamma_R$)



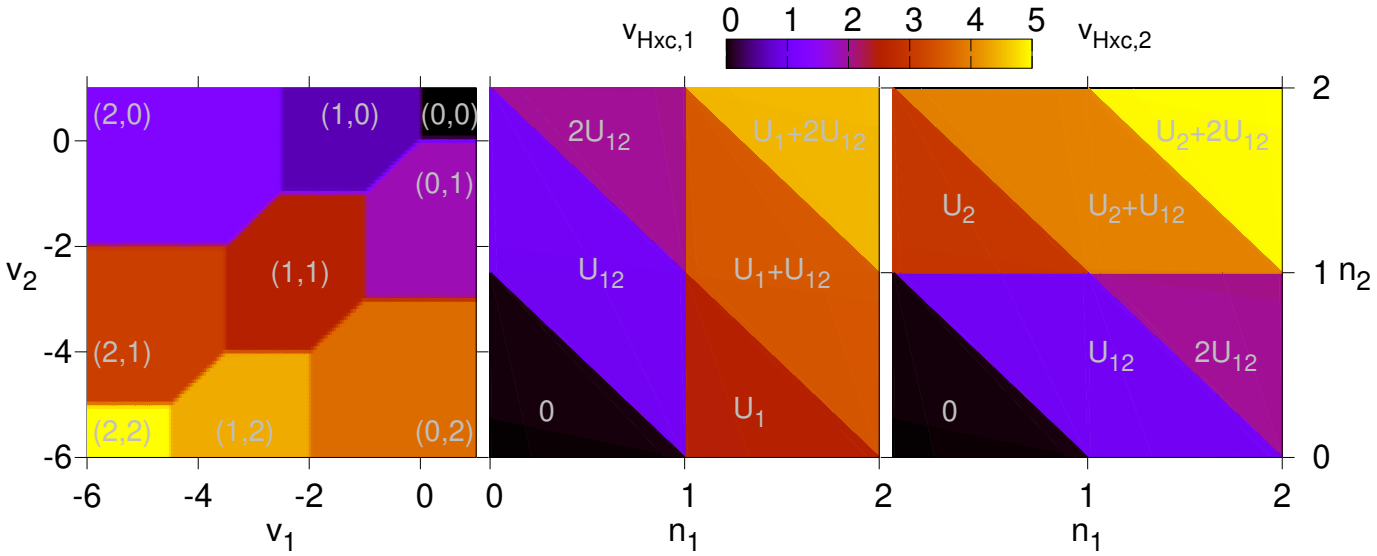
- again smeared steps of height $U/2$ (U) for Hxc gate (xc bias) with edges at piecewise linear functions of $\Delta_K^{(\pm)}(N, I)$
- complex pattern of vertices in (N, I)-plane simplifies in asymmetric limit

a), d) $\gamma_L/\gamma = 0.5$; b), e) $\gamma_L/\gamma = 0.25$; c), f) $\gamma_L/\gamma = 5 \times 10^{-5}$;

Digression: Stability diagrams and Hxc potentials

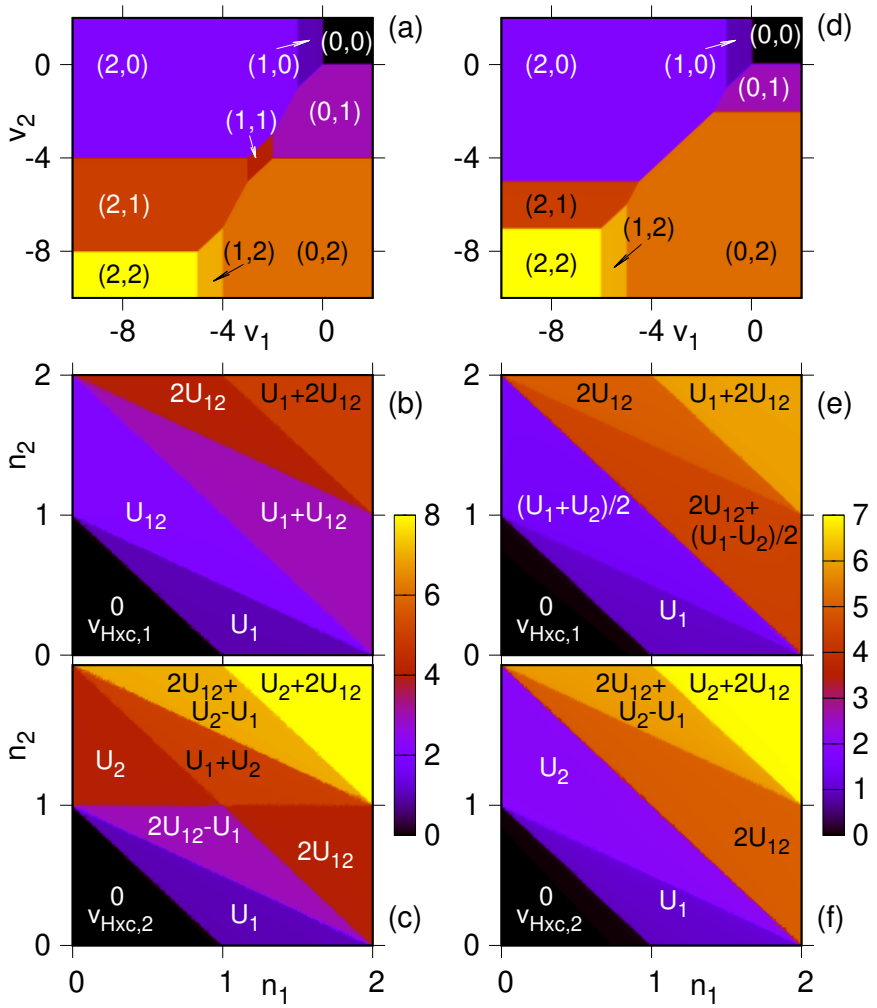
Hxc potentials of uncontacted double dot at equilibrium exhibit steps in low temperature limit depending on parameter regimes; Stability diagrams and Hxc potentials are intimately connected

Ex: Regime I: $U_i > U_{12}$



GS occupations (integers) correspond to vertices in $n_1 - n_2$ plane regions for two different vertices touch $\longrightarrow$ step in Hxc potential, step height equals length of touching line in stability diagram

Digression: Stability diagrams and Hxc potentials (cont.)



Left column: Regime II:

$U_1 < U_{12} < \bar{U}$

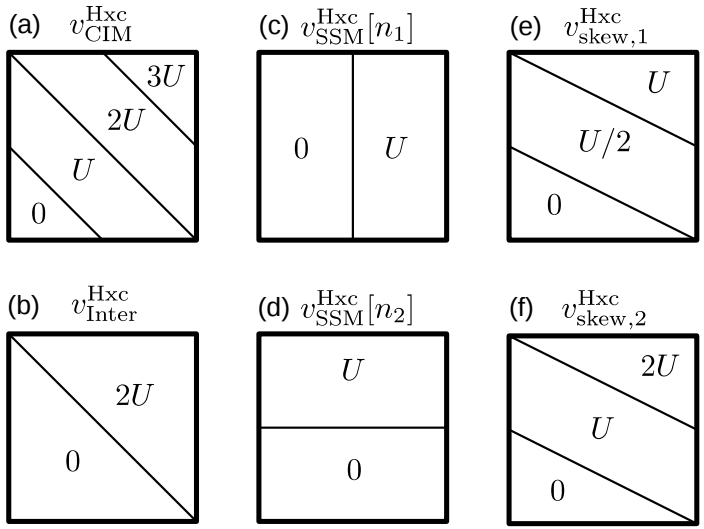
where $\bar{U} = (U_1 + U_2)/2$

note presence of steps along lines from $(0,1)$ to $(2,0)$ and from $(0,2)$ to $(2,1)$

Right column: Regime III:

$U_1 \leq \bar{U} \leq U_{12}$

Digression: Building blocks for Hxc potentials



Construct Hxc ptls by adding building blocks: steps at diff. positions in $n_1 - n_2$ plane

$v^{\mathrm{Hxc}}_{\mathrm{CIM}}(U)[N]$: Hxc ptl. of Constant Interaction Model

$v^{\mathrm{Hxc}}_{\mathrm{inter}}(U)[N]$: Hxc ptl. due to inter-Coulomb repulsion U_{12}

Example: Hxc potential for Regime I

$$v^{\alpha}_{\mathrm{Hxc}}[n_1, n_2] = v^{\mathrm{CIM}}_{\mathrm{Hxc}}(U_{12})[N] + v^{\mathrm{SIAM}}_{\mathrm{Hxc}}(U_\alpha - U_{12})[n_\alpha]$$

with $N = n_1 + n_2$

Reference: PRB **102**, 035159 (2020)

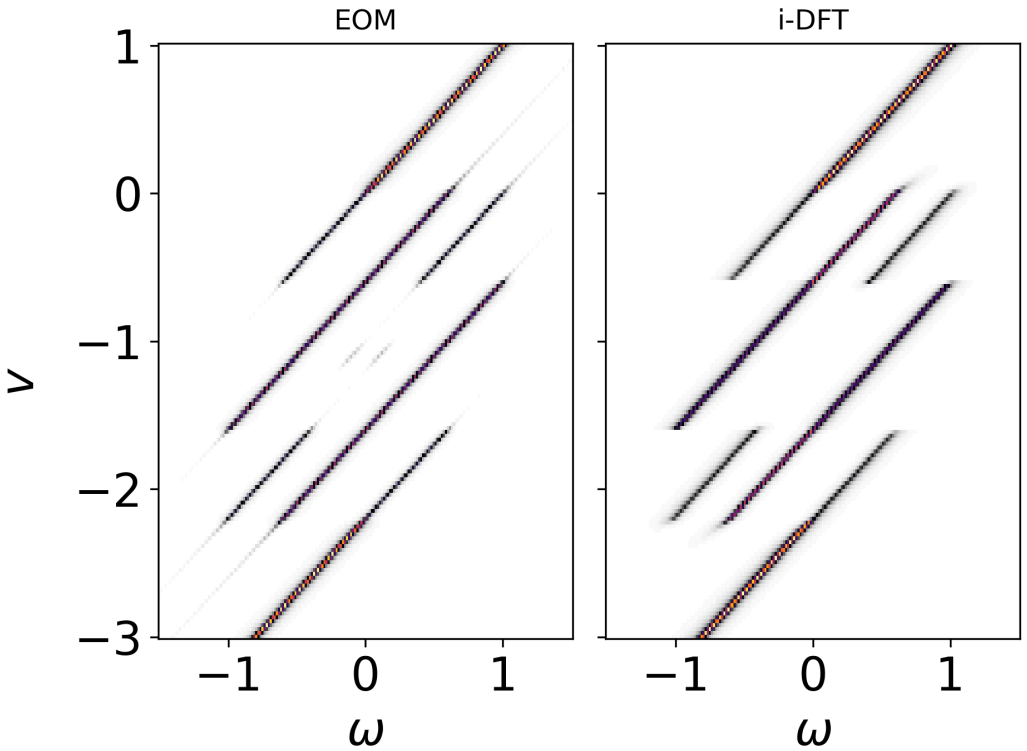
Construction of xc bias for double dot in STM limit

use same idea of constructing $V_{\rm xc}^{\alpha}[n_1, n_2, I]$ (for only dot α being connected to STM tip) as sum of building blocks, e.g.,

xc bias for Regime I

$$V_{\rm xc}^{\alpha}[n_1, n_2, I] = V_{\rm xc}^{\rm CIM}(U_{12})[N, I] + V_{\rm xc}^{\rm SIAM}(U_{\alpha} - U_{12})[n_{\alpha}, I]$$

Total spectral functions for Regime I



total spectral functions as function of frequency ω and average on-site potential $v = (v_1 + v_2)/2$

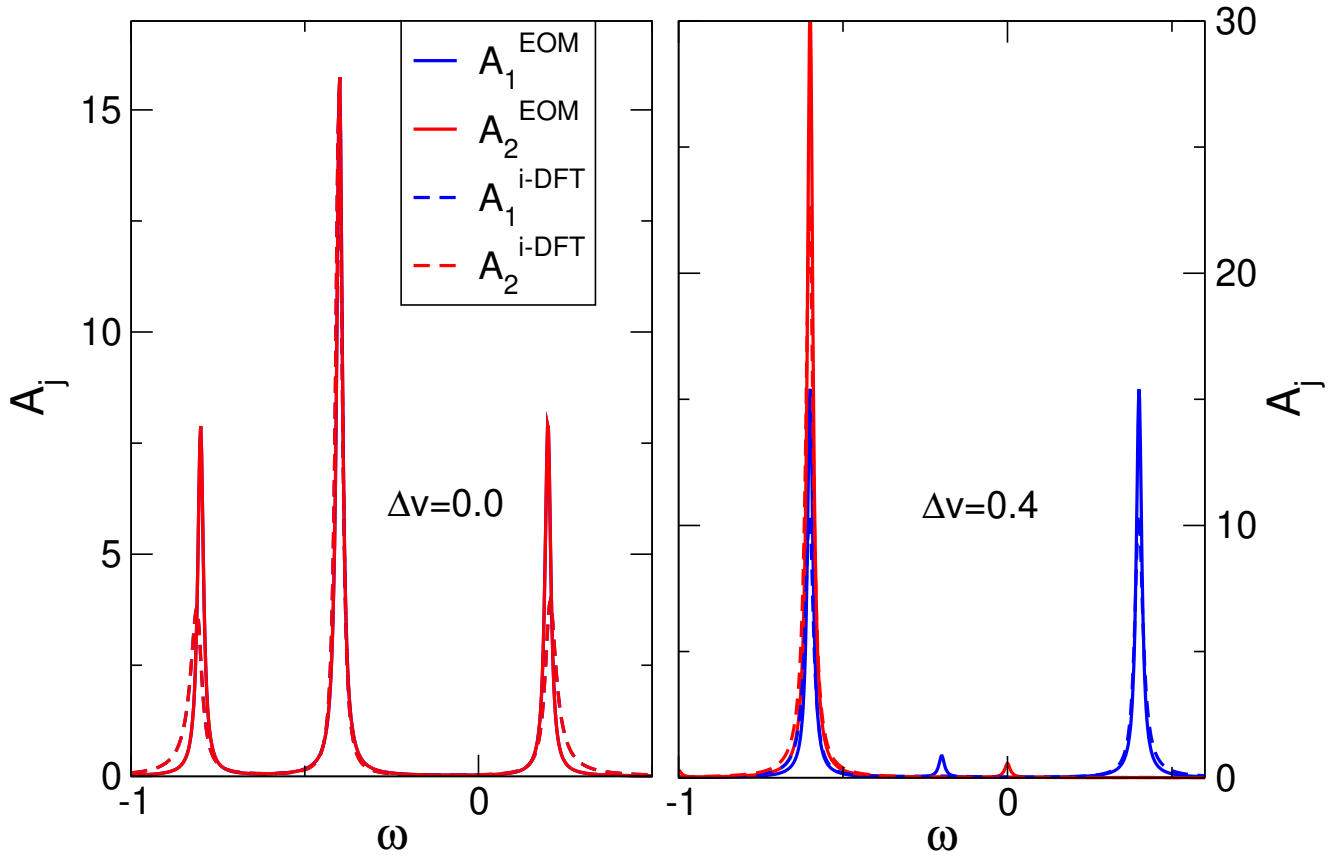
parameters:

$U_1 = U_2 = 1.0, U_{12} = 0.6$

$\gamma = 0.02, \Delta v = v_1 - v_2 = 0$

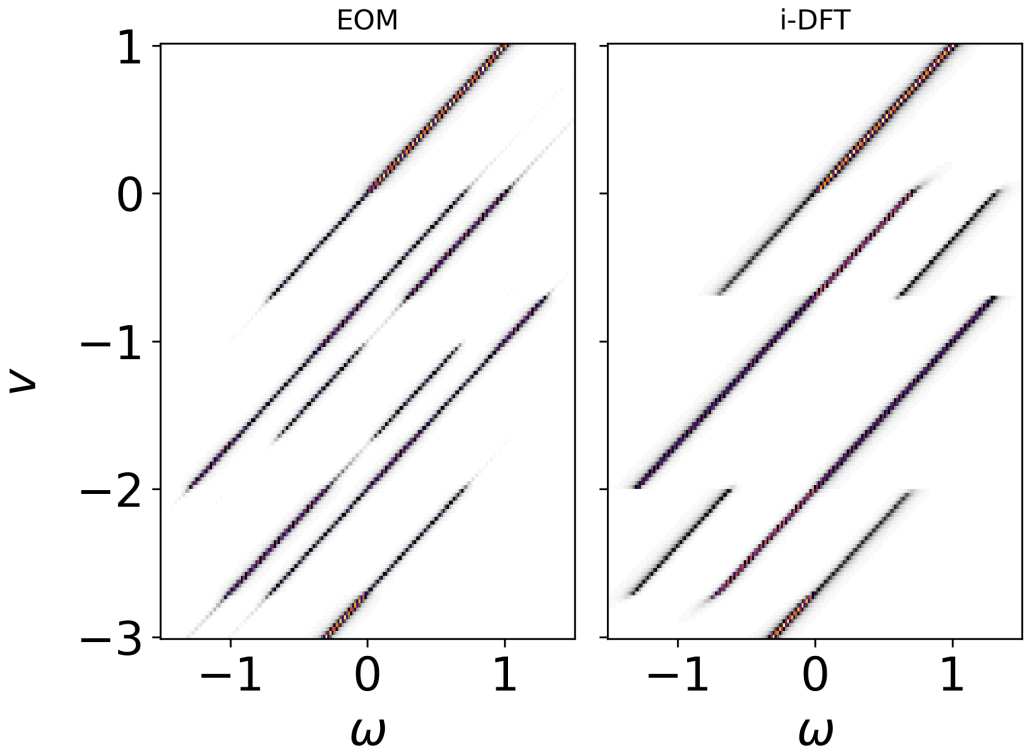
comparison to results from Equations of Motion (EOM) for Green functions with truncation scheme of PRB **111**, 115108 (2025)

Local spectral functions for Regime I



local spectral functions for same parameters for $v = -2$ and $\Delta v = 0$ and $\Delta v = 0.4$

Total spectral functions for Regime III: not all is well



total spectral functions as function of frequency ω and average on-site potential v

parameters:

$U_1 = U_2 = 0.7, U_{12} = 1.0$

$\gamma = 0.02, \Delta v = 0$

i-DFT xc potentials: Inclusion of Kondo effect

i-DFT expression for zero-bias conductance: linearize i-DFT eq. for current $\longrightarrow$

exact expression for zero-bias conductance

$$G = \frac{G_s}{1 - G_s \left.\frac{\partial V_{\rm xc}}{\partial I}\right|_{I=0}}$$

note: the KS zero-bias conductance G_s already accounts for the Kondo effect (at $T = 0$ if accurate ground state functional is used) $\longrightarrow$ in order to incorporate Kondo physics in our functional, make sure that correction term in denominator vanishes

i-DFT xc potentials: inclusion of Kondo effect

in ideal STM limit, modify xc bias for only site j connected to tip to include Kondo physics

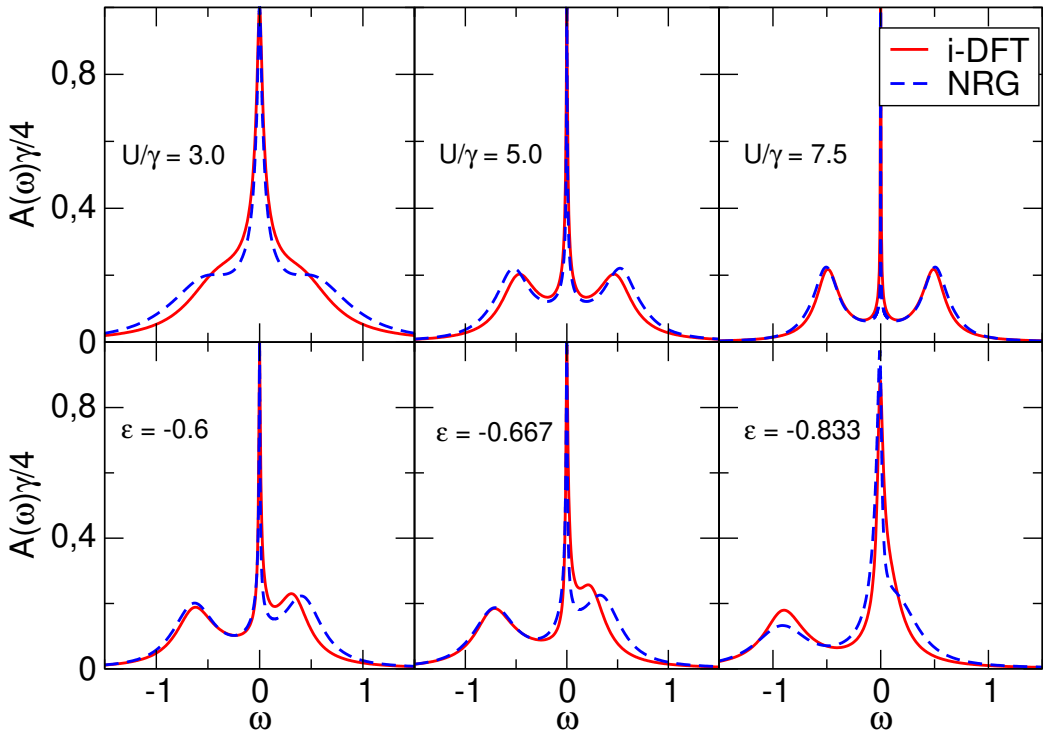
parametrization of xc bias including Kondo effect

$$V_{\mathrm{xc,j}}[n_1, n_2, I] = (1 - a[I])\, \bar{V}_{\mathrm{xc}}[n_1, n_2, I]$$

$$a[I] = 1 - \frac{2}{\pi}\mathrm{atan}\left[\lambda\left(\frac{I}{4W_j\gamma_T}\right)^2\right]$$

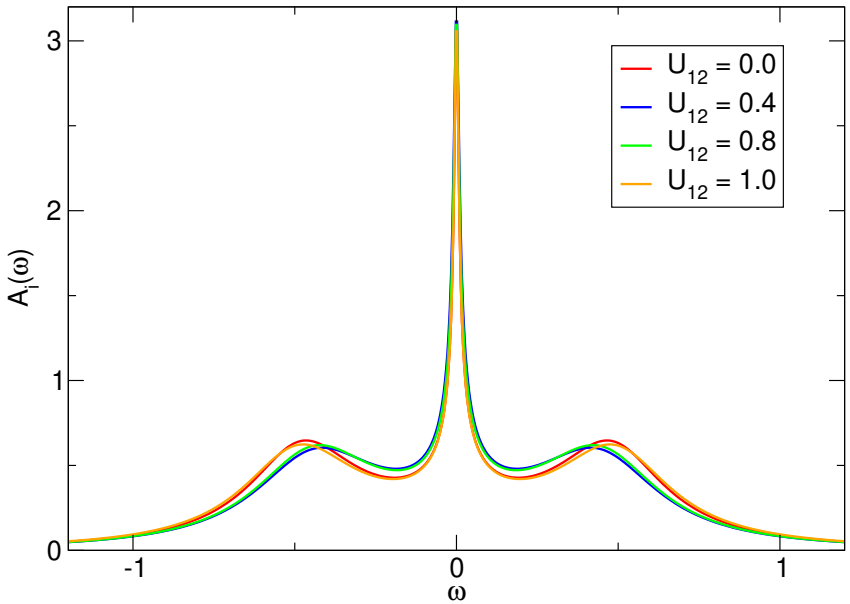
with parameter $\lambda = 0.16$ and $W_j = 0.16\gamma/U_j$

i-DFT spectral functions for SIAM

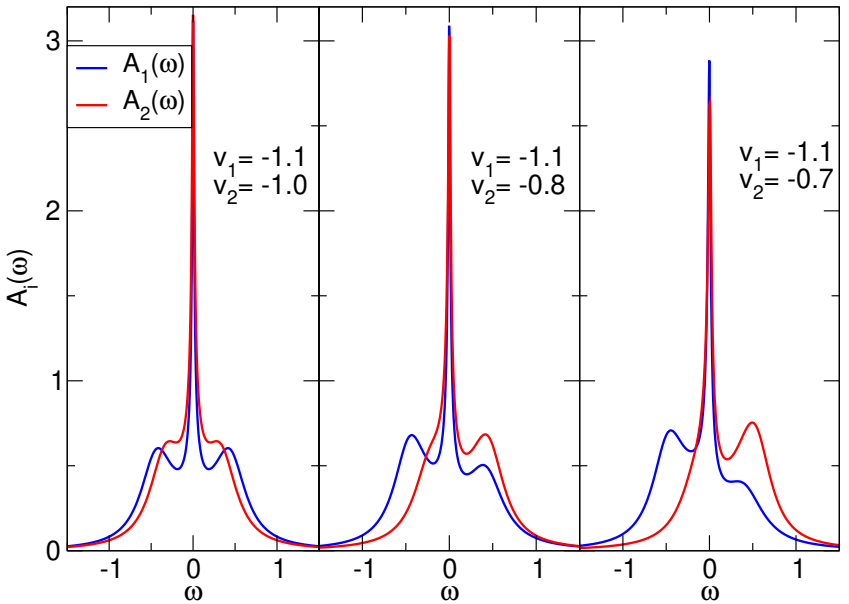


upper panels: at particle-hole symmetry for different U/γ
lower panels: at $U/\gamma = 5$ for different on-site energies ε
blue: NRG results (Motahari et al, PRB **94**, 235133 (2016))

i-DFT spectral functions (Reg.I) including Kondo effect

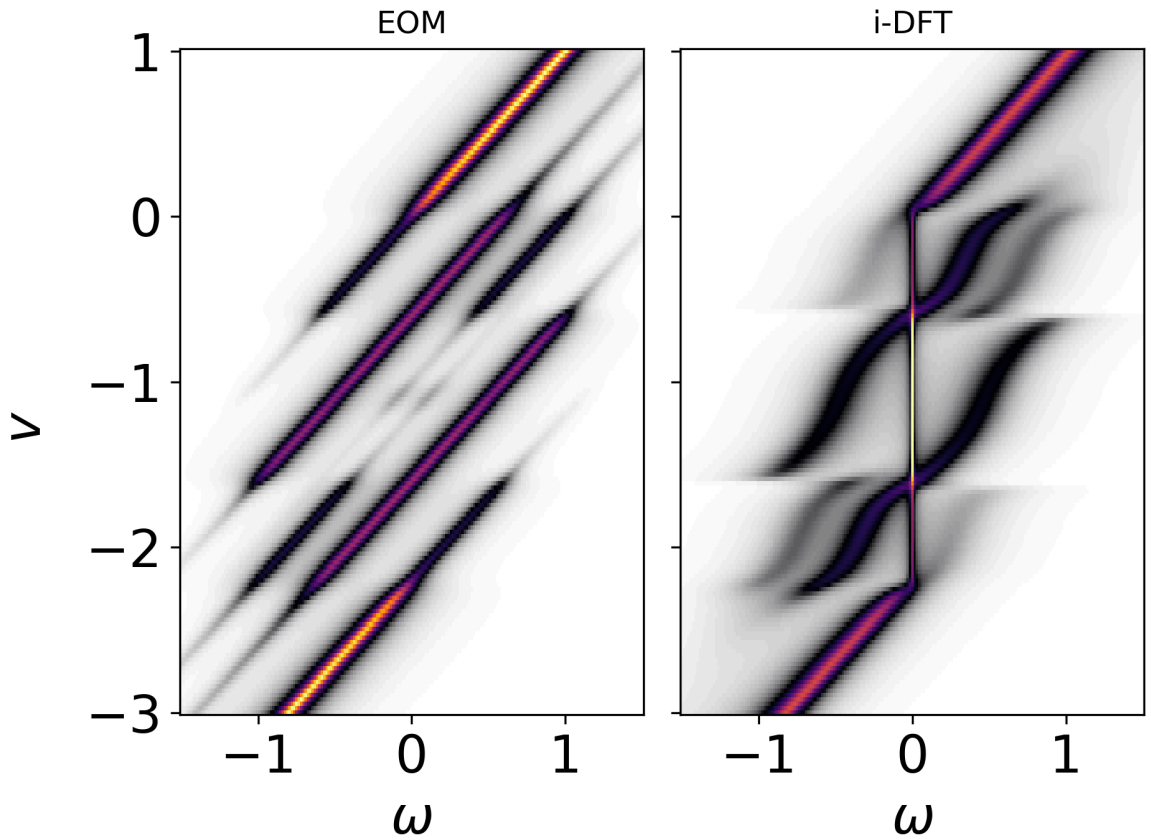


$U_1 = U_2 = 1.0$ for different U_{12} at ph symmetry ($\gamma = 0.2$)



$U_1 = 1.0, U_2 = 0.8, U_{12} = 0.6$ at different on-site energies ($\gamma = 0.2$)

i-DFT spectral functions (Reg.I) including Kondo effect



EOM and i-DFT spectral functions for $U_1 = U_2 = 1.0, U_{12} = 0.6$ and $\gamma = 0.1$ with i-DFT including the Kondo effect

Mott Metal-Insulator transition from i-DFT

Reference:

- D. Jacob, G. Stefanucci, S. Kurth, Phys. Rev. Lett. **125**, 216401 (2020)

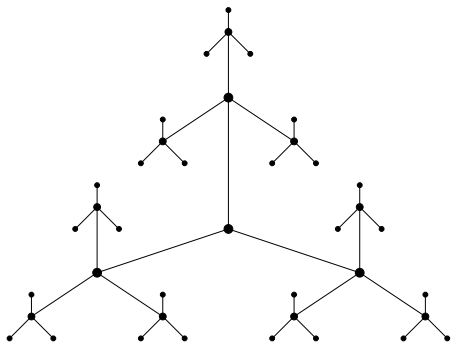
Hubbard model on various lattices

Hamiltonian of Hubbard model

$$\hat{H} = -\sum_{\sigma} \sum_{<i,j>} t(\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + \text{H.c.}) + \sum_{\sigma}\sum_{i} v_i \hat{n}_i + \sum_{i} U \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$

where the sum $< i, j >$ is over the nearest neighbors of the lattice
here: look at uniform systems $v_i = v$ for simple cubic and Bethe lattice

example:
Bethe lattice at coordination $M = 3$



M=3

Conditions on xc bias from many-body approach

in DMFT: many-body self energy $\Sigma(\omega)$ becomes local (independent of $\mathbf{k}$ in momentum space) $\longrightarrow$ derive relations between MB and KS spectral functions at $\omega = 0$ with quasiparticle weight Z

$$A(0) = A_s(0) \quad A'(0) = Z^{-1} A_s'(0) \quad A''(0) = Z^{-2} A_s''(0)$$

can be used to deduce following conditions (2)-(4) on xc bias

Conditions on xc bias

(1) $V_{\rm xc}[\tilde{I} = 0] = 0$

(2) $\frac{\partial V_{\rm xc}}{\partial \tilde{I}}\big|_{\tilde{I}=0} = 0$

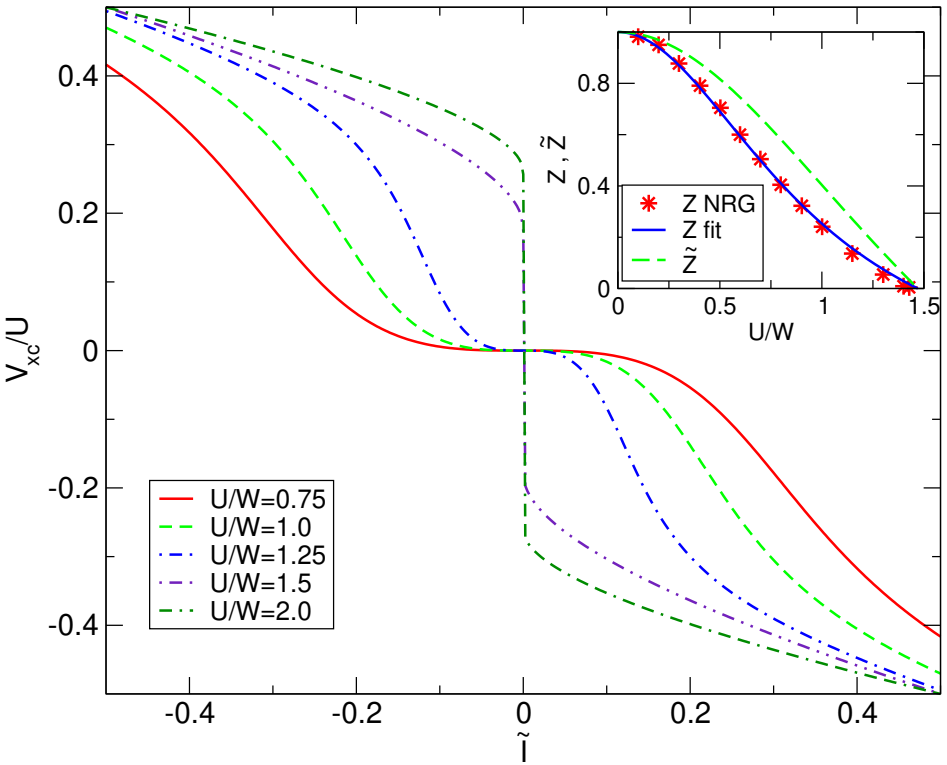
(3) $\frac{\partial^2 V_{\rm xc}}{\partial \tilde{I}^2}\big|_{\tilde{I}=0} = (Z^{-1} - 1)\frac{A_s'(0)}{(A_s(0))^3}$

(4) $\frac{\partial^3 V_{\rm xc}}{\partial \tilde{I}^3}\big|_{\tilde{I}=0} = \frac{(Z^{-2}-1)}{(A_s(0))^4}\left(A_s''(0) - 3\frac{(A_s'(0))^3}{A_s(0)}\right)$

where $\tilde{I} = I/(2\gamma)$

Construction of xc bias

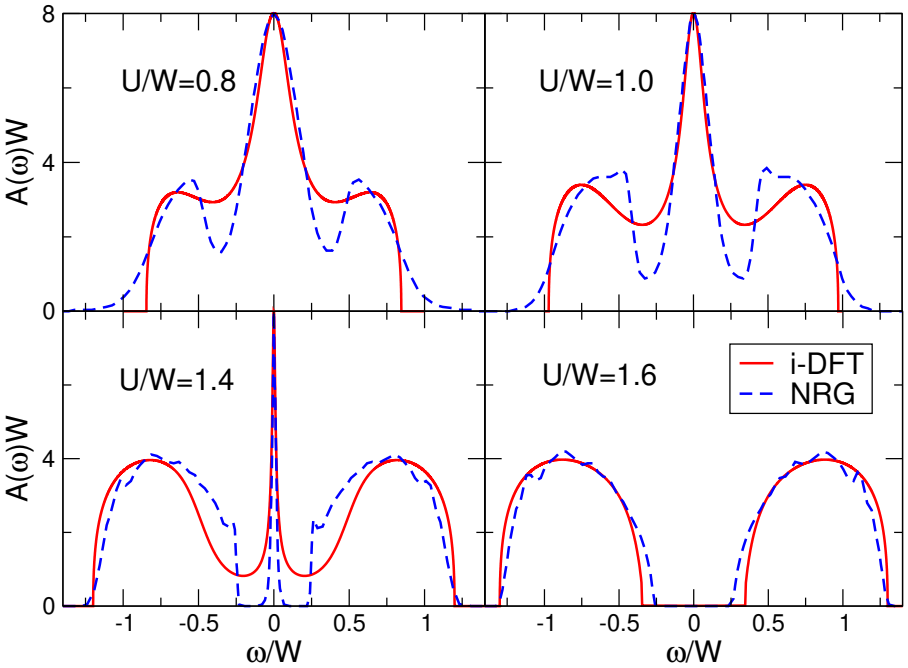
xc bias and quasi-particle weight Z (from Bulla, PRL **83**, 136 (1999)) for different values of U for Bethe lattice at infinite coordination (W: bandwidth at $U = 0$)



note that the step is suppressed (condition (2)) for small U (in the metallic phase)

i-DFT spectra of Hubbard model on different lattices

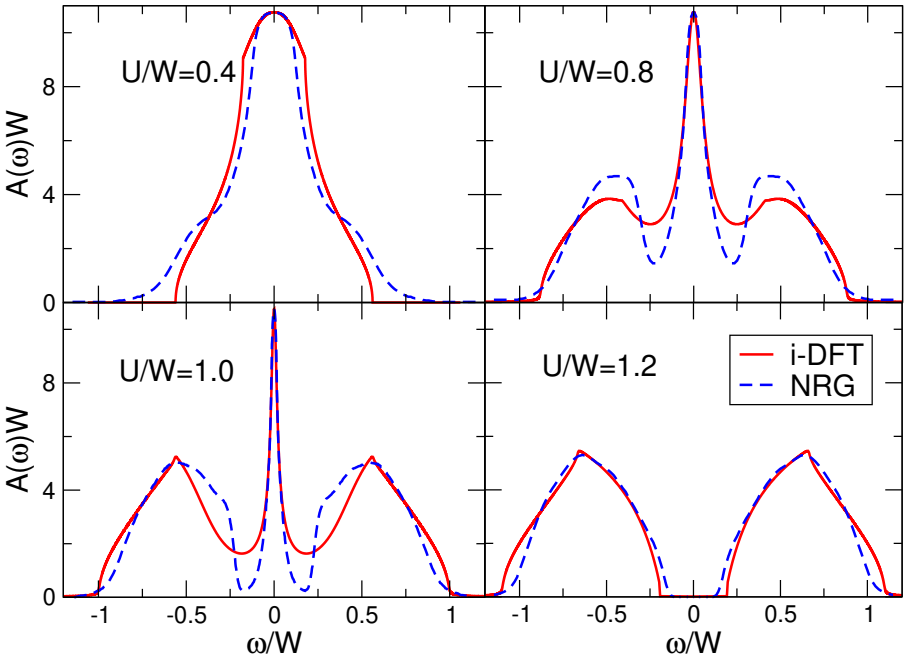
Spectral function of Hubbard model on Bethe lattice from i-DFT compared to NRG



NRG data from Zitko et al, PRB **79**, 085106 (2009). Note the disappearance of the quasiparticle peak as U increases (metal-insulator transition).

i-DFT spectra of Hubbard model on different lattices

Spectral function of Hubbard model on simple cubic lattice from i-DFT compared to NRG



NRG data from Zitko et al, PRB **80**, 245112 (2009). Again: Mott metal-insulator transition captured by i-DFT.

Summary

- Many-body spectral functions from differential conductance in "STM limit" (at $T = 0$)
- i-DFT gives relation between KS and many-body spectral function
- xc bias functionals for model systems: the work is in the Coulomb blockade part of the xc bias; inclusion of Kondo effect straightforward
- Mott metal-insulator transition can be described by i-DFT

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